Density matrix formulation of dynamical systems

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Density matrix formulation of dynamical systems

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Physical systems that are dissipating, mixing, and developing turbulence also irreversibly transport statistical density. However, predicting the evolution of density and entropy from atomic and molecular scale dynamics is challenging for non-steady, open, and driven nonequilibrium processes. Here, we establish a theory to address this challenge for classical dynamical systems that is analogous to the density matrix formulation of quantum mechanics. We show that a classical density matrix is similar to the phase-space metric and evolves in time according to generalizations of Liouville's theorem and Liouville's equation for non-Hamiltonian systems. The traditional Liouvillian forms are recovered in the absence of dissipation or driving, by imposing trace preservation or by considering Hamiltonian dynamics. Local measures of dynamical instability and chaos are embedded in classical commutators and anti-commutators and directly related to Poisson brackets when the dynamics are Hamiltonian. Because the classical density matrix is built from the Lyapunov vectors that underlie classical chaos, it offers an alternative computationally-tractable basis for the statistical mechanics of nonequilibrium processes that applies to systems that are driven, transient, dissipative, regular, and chaotic.

I. INTRODUCTION

Whether classical or quantum mechanical, the transport of statistical density is our primary means of making statistical predictions of macroscopic behavior from microscopic dynamics [1]. Classically, Jacobi's form of Liouville's equation of motion for the phase space density of mechanical systems is the foundation of statistical mechanics [2]. Through extensive efforts, it has many forms and approximations, including the Boltzmann equation, the Vlasov approximation, the Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy, that underlie applications across physics and chemistry [1]. These various mathematical forms lead to macroscopic predictions with varying fidelity and numerical tractability. In quantum mechanics, the Liouville-von Neumann equation describes the evolution of the density operator [3]; it is the fundamental equation of quantum statistical mechanics and a main ingredient in quantum computing, tomography, and decoherence [4]. To translate between the classical and quantum mechanical Liouville equations, one can use Dirac's rule [5] of replacing Poisson brackets by commutators. Here, we establish a density matrix formalism for classical systems that supplants Dirac's heuristic with a more direct correspondence between these physical theories.

There are other classical theories that add weight to the question of whether formulations of quantum mechanics might have classical counterparts that could advance statistical physics [6]. Operator-theoretic methods, such as Frobenius-Perron and its dual Koopman formalism [7], give a formal analogy to quantum mechanics by lifting the description of classical systems to infinite dimensions [8]. They preserve global nonlinear features and guarantee exact linearization of the dynamics, providing useful connections between classical dynamical systems and statistical physics [9, 10]. However, they can be difficult to apply to systems under active external control and to find observables representing the nonlinear system in the lifted linear space [11]. Symmetries can make the calculation of the Koopman operator approximation and its spectral properties more efficient [12], but, in practice [13, 14], the number of variables must be truncated to finite-dimensions (e.g., through extended [15, 16] or kernel [17] dynamic mode decomposition [18]).

While Liouville's equation is the formal foundation of nonequilibrium statistical physics, many theories avoid, approximate, or subject it to model specific solutions [1]. Here, we construct a classical density matrix formulation of dynamical systems on the local stability of nonlinear dynamics [20] – Lyapunov exponents and vectors [21]. The infinitesimal perturbations, Lyapunov vectors, defining the density matrix have been used to analyze rare trajectories [22], jamming [23], nonequilibrium self-assembly [24], equilibrium and nonequilibrium fluids [25–27], and critical phenomena [28]. From these finite-dimensional vectors, we derive a classical analogue of the von Neumann equation for the density matrix dynamics. We show this classical density matrix is similar to the (dual) metric tensor and that its determinant evolves according to a generalized Liouville equation and satisfies a generalized Liouville theorem. And, imposing a norm-preserving dynamics with Lyapunov exponents not only normalizes the density matrix, it reinstates the form of the usual Liouville equation for generic, non-Hamiltonian dynamical systems.

To start, we define an unnormalized density matrix from the linearization of the classical dynamics, Sec. II. The properties of this density matrix lead to a generalization of Liouville's theorem and equation, Sec. II A. For Hamiltonian dynamics, Sec. II C, we show the reduction

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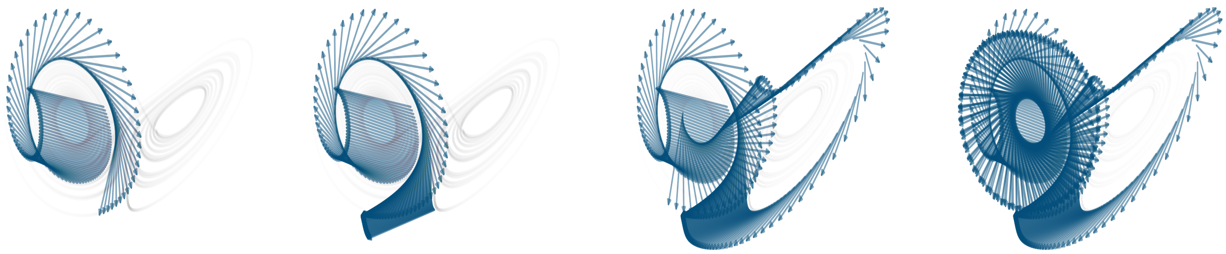

FIG. 1. Snapshots of the strange attractor and an initially random, unit perturbation transported along a chaotic solution of the Lorenz-Fetter model. Parameters are those originally used by Lorenz [19]: $\mu = 10$, $\beta = 8/3$, $\rho = 28$.

to the usual Liouville theorem and equation and establish a connection to Poisson brackets. The dynamics of a normalized density matrix, Sec. III, transform the generalized Liouville's theorem and equation to the usual form, Sec. III A. In Sec. III B, we discuss a basis representation of the density matrix, a common consideration in quantum mechanics, with another form of Liouville's theorem.

II. DYNAMICS OF THE CLASSICAL UNNORMALIZED DENSITY MATRIX

Consider a classical dynamical system with state-space variables $\{x^i\}$. At any moment in time, these variables together mark a point $\boldsymbol{x}(t) := [x^1(t), x^2(t), \ldots, x^n(t)]^\top$ in an n-dimensional state space $\mathcal{M}$ that evolves according to: $\dot{\boldsymbol{x}} = \boldsymbol{F}[\boldsymbol{x}(t)]$. Perturbations to the system will also evolve under the flow of the dynamics. Because of their analytical and computational tractability, infinitesimal perturbations $|\delta\boldsymbol{x}(t)\rangle := [\delta x^1(t), \delta x^2(t), \ldots, \delta x^n(t)]^\top \in T\mathcal{M}$ and their linearized dynamics are a well established means of analyzing the stability of nonlinear dynamical systems [21]. These perturbations to the initial condition stretch, contract, and rotate over time,

$$|\delta\dot{\boldsymbol{x}}(t)\rangle = \boldsymbol{A}[\boldsymbol{x}(t)]\,|\delta\boldsymbol{x}(t)\rangle\,, \tag{1}$$

as they evolve with the phase point under the local stability matrix $\boldsymbol{A} := \boldsymbol{A}[\boldsymbol{x}(t)] = \boldsymbol{\nabla F}$ with elements $(\boldsymbol{A})^i_j = \partial\dot{x}^i(t)/\partial x^j(t)$. Figure 1 shows a unit perturbation vector as it is transported across the Lorenz attractor.

A common approach is to consider an infinitesimal k-dimensional phase space volume surrounding the phase point $\boldsymbol{x}(t)$ that transforms its shape over time. We take the volume to be spanned by a finite set $\{|\delta\boldsymbol{\psi}_i\rangle\}$ of $0 < k \leq n$ linearly independent tangent vectors [21], $|\delta\boldsymbol{\psi}_i\rangle \in T\mathcal{M}$ with $i = 1, 2, \ldots, k$, that also obey the linearized dynamics. Examples include Gram-Schmidt and covariant Lyapunov vectors [29, 30]. For the present discussion, we assume the elements of tangent vectors (represented by a ket/column or a bra/row) are real. If the dynamics are Hamiltonian, then according to Liouville's theorem, the volume spanned by n of these tangent vectors is conserved.

Compared to quantum mechanics, Eq. 1 is analogous to Schrödinger's equation [5]. The difference is that instead of infinite-dimensional, complex Hilbert space vectors, here we are considering classical, finite-dimensional, and real tangent-space vectors. Continuing this analogy, we can realize that an alternative representation of quantum states is the density operator [4, 31, 32]. The quantum density operator is used in quantum technology and statistical mechanics and particularly important for many-body and open quantum systems [33]. Because of the widespread use of this formulation in quantum mechanics and the need for a statistical-mechanical theory for open, driven classical systems, we break from traditional classical dynamical systems by defining the classical density matrix:

$$\boldsymbol{\xi}(t) := \sum_{i=1}^{k} |\delta\boldsymbol{\psi}_i(t)\rangle\langle\delta\boldsymbol{\psi}_i(t)|\,. \tag{2}$$

Expressed using tangent vectors $|\delta\boldsymbol{\psi}_i\rangle$, the density matrix is the outer product of tangent vectors (or what Gibbs called the dyadic product [34]). This *unnormalized* matrix represents an alternative state of a classical dynamical system at a phase space point. To our knowledge, this classical density matrix has not been defined previously. For classical many-body systems in position-momentum phase space, $\boldsymbol{x} = (\boldsymbol{q}, \boldsymbol{p})$, it is a mechanical function of perturbations to positions and momenta $\delta\boldsymbol{\psi}_i = (\delta\boldsymbol{q}^i, \delta\boldsymbol{p}_i)$. For example, choosing the orthogonal tangent vectors $|\delta\boldsymbol{\psi}_1\rangle = \sqrt{\delta q\delta p}(1,1)^\top$ and $|\delta\boldsymbol{\psi}_2\rangle = \sqrt{\delta q\delta p}(1,-1)^\top$, the density matrix of any Hamiltonian system with one degree of freedom (e.g., the harmonic oscillator) is $\boldsymbol{\xi} = \delta q\delta p\,\mathbb{1}_{2\times 2}$, where $\mathbb{1}_{n\times n}$ is the $n \times n$ identity matrix.

The dynamics of the classical density matrix involve a classical commutator and anti-commutator. Partitioning the stability matrix $\boldsymbol{A} = \boldsymbol{A}_+ + \boldsymbol{A}_-$ into its symmetric and anti-symmetric parts, $\boldsymbol{A}_\pm = \frac{1}{2}(\boldsymbol{A} \pm \boldsymbol{A}^\top)$, the time evolution of $\boldsymbol{\xi}$,

$$\frac{d\boldsymbol{\xi}}{dt} = \{\boldsymbol{A}_+, \boldsymbol{\xi}\} + [\boldsymbol{A}_-, \boldsymbol{\xi}], \tag{3}$$

is a purely classical analogue of the Liouville-von Neumann equation in quantum dynamics. Its solution,

$$\boldsymbol{\xi}(t) = \boldsymbol{M}(t,t_0)\boldsymbol{\xi}(t_0)\boldsymbol{M}^\top(t,t_0), \tag{4}$$

is in terms of the propagator, $(\boldsymbol{M})^i_j = \partial x^i(t)/\partial x^j(t_0)$ when $\boldsymbol{\xi}$ is built from the tangent vectors $\{|\delta \boldsymbol{x}^i\rangle\}$ evolved by $\boldsymbol{M}$, App. A. While it is generally non-symmetric, the stability matrix $\boldsymbol{A}$ plays the role of the quantum mechanical Hamiltonian in the *classical* commutator $[\boldsymbol{X}, \boldsymbol{Y}] = \boldsymbol{Y}\boldsymbol{X} - \boldsymbol{X}\boldsymbol{Y}$ and anti-commutator $\{\boldsymbol{X}, \boldsymbol{Y}\} = \boldsymbol{Y}\boldsymbol{X} + \boldsymbol{X}\boldsymbol{Y}$.

What stands out about the evolution of the density matrix is that its dynamics are entirely computable from standard methods in dynamical systems theory (viz., Lyapunov vectors) [21]. Figure 1 shows the evolution of a normalized Lyapunov vector on the Lorenz attractor that follows Eq. 3. This classical density gives a geometric representation of deterministic, mechanical systems that leads to a generalized form of Liouville's theorem and equation governing the dynamics of compressible phase space volumes. While the dynamics of the harmonic oscillator are conservative, the connection between the density matrix and phase space volume is clear: the density matrix $\boldsymbol{\xi} = \delta q \delta p\, \mathbb{1}_{2\times 2}$ has a trace, $2\delta q\delta p$, and determinant, $(\delta q \delta p)^2$, that are directly related to the phase space volume, $d\mathcal{V} = \delta q \delta p$.

A. Generalization of Liouville's theorem

From the equation of motion for the classical density matrix, we can show the determinant of $\boldsymbol{\xi}$ is directly related to both Liouville's equation and theorem. Liouville's theorem and equation are the foundation for nonequilibrium statistical mechanics [1, 9, 10] and the point at which statistical mechanics departs from classical Hamiltonian dynamics. However, the density matrix here leads to generalized forms of Liouville's theorem and equation that hold for non-Hamiltonian systems, systems that may be open, closed, passive, or driven.

To establish this connection between the density matrix and a generalized version Liouville's theorem, consider a complete set of linearly independent tangent vectors $\{\delta\boldsymbol{\psi}_i\}$. The set spans the entire n-dimensional phase space volume, $d\mathcal{V}$, and with an associated unnormalized density matrix $\boldsymbol{\xi}$ (i.e., $k = n$ in Eq. 2). Regardless of the state space variables, the (square of the) phase space volume is determined by the determinant $|\boldsymbol{\xi}|$, which has the equation of motion (App. B):

$$\frac{1}{2}\frac{d}{dt}\ln|\boldsymbol{\xi}(t)| = \boldsymbol{\nabla}\cdot\dot{\boldsymbol{x}} = \mathrm{Tr}\,\boldsymbol{A}_+ = \Lambda. \tag{5}$$

Both this equation of motion and its solution,

$$|\boldsymbol{\xi}(t)| = |\boldsymbol{\xi}(t_0)|\, e^{2\int_{t_0}^{t}\Lambda(t')\,dt'}, \tag{6}$$

depend on the divergence of the phase space velocity $\dot{\boldsymbol{x}}$ or the phase space volume contraction/expansion rate $\Lambda = \mathrm{Tr}\,\boldsymbol{A}_+ = \mathrm{Tr}\,\boldsymbol{A}$. The determinant $|\boldsymbol{\xi}|$ of the density matrix, which is a potentially mechanical function, has an equation of motion that is similar to the equation of motion for the statistical density [35]. In both of these equations, the phase space contraction rate Λ is the sum of the Lyapunov exponents [9], which can be related to physical quantities. For example, the phase space contraction rate is related to the entropy flow rate in fluid transport [9]. It is also related to the thermodynamic dissipation for systems in nonequilibrium steady states, provided the dynamics are subject to a deterministic thermostat [36].

Now, we can identify the absolute value of the density matrix determinant as the volume of an element of state space, $|\boldsymbol{\xi}|^{1/2} = d\mathcal{V}$. Because the density matrix is well defined for physical systems that are open, both at the microscopic and macroscopic levels, their phase space volume element need not remain conserved with time and will generally evolve in time according to Eq. 6. Traditionally, the geometric interpretation of Liouville's theorem is that the velocity field $\dot{\boldsymbol{x}}$ has zero divergence: $\boldsymbol{\nabla}\cdot\dot{\boldsymbol{x}} = \mathrm{Tr}(\boldsymbol{\mathcal{H}}) = 0$. As a consequence, the "phase fluid" flow is incompressible, phase space volumes are conserved $d\mathcal{V}(t) = d\mathcal{V}(t_0)$, and $\Lambda = 0$. That is, from the determinant, we can see Liouville's theorem and equation from the stability matrix $\boldsymbol{\mathcal{H}}$ for a dynamics with Hamiltonian H. However, in dissipative systems we must account for this "compressibility" of the state-space volume, $d\mathcal{V}(t) \neq d\mathcal{V}(t_0)$. This observation suggests that Eq. 6 might lead to a generalization of Liouville's theorem for non-Hamiltonian systems.

The volume element $d\mathcal{V}(t)$ spanned by a set of basis vectors has a coordinate transformation: $d\mathcal{V}(t) = |\boldsymbol{M}(t,t_0)|\, d\mathcal{V}(t_0)$. Combining this fact with the determinant of Eq. 4, we obtain a generalization of Liouville's theorem in terms of the classical density matrix:

$$|\boldsymbol{\xi}(t)|^{-\frac{1}{2}}\,d\mathcal{V}(t) = |\boldsymbol{\xi}(t_0)|^{-\frac{1}{2}}\,d\mathcal{V}(t_0). \tag{7}$$

Any dynamics conserves the measure, $|\boldsymbol{\xi}|^{-\frac{1}{2}}d\mathcal{V}$ or $e^{-\int_{t_0}^{t}\Lambda(t')\,dt'}d\mathcal{V}$. For dissipative systems with $\Lambda < 0$, volumes contract at a rate $\mathrm{Tr}\,\boldsymbol{A}$. In the Lorenz-Fetter model, for example, $\boldsymbol{A}$ is constant, so $|\boldsymbol{\xi}|$ decays linearly on a semi-log scale with a slope proportional to $2\,\mathrm{Tr}\,\boldsymbol{A}$ as shown in Fig. 2. Dynamical systems that are open, exchanging matter or energy with their environment, or driven by external fields, will have a density matrix that varies in time and a determinant that satisfies this version of Liouville's equation. When the dynamics are Hamiltonian, we recover the conventional form of Liouville's theorem for phase space volumes [2] because $\Lambda = 0$. That is, for Hamiltonian dynamics, the determinant of the unnormalized density matrix, $|\boldsymbol{\xi}|$, is a constant of motion.

With the determinant of the density matrix giving Liouville's theorem, we can consider exactly how this matrix is related to the geometry of phase space. The connection comes from Riemannian geometry. Equation 7 is the transformation of a metric determinant on a Riemannian manifold of an arbitrary curvature and endowed with a covariant metric tensor $\boldsymbol{g}_{\boldsymbol{\xi}}$. So, we would expect the density matrix to be related to this metric tensor. As we show in Appendix B, the determinant $|\boldsymbol{\xi}|$ is equal to the metric determinant, and the inverse of the unnormalized density matrix, $\boldsymbol{\xi}^{-1}$, is similar to the (covariant) metric tensor, $\boldsymbol{g_\xi}$.

Metrics have been considered previously in nonlinear dynamics and the statistical mechanics of non-Hamiltonian systems [37–42]. However, these previous approaches do not involve the classical density matrix we define here. Instead, they require finding the metric factor (the square root of the metric determinant) by solving a generalized Liouville's equation [42], which can be challenging. Again, this metric factor is used to define a volume form, which is invariant under the compressible flow. Here, because the inverse of the determinant of the classical density matrix $|\boldsymbol{\xi}|^{-1}$ is equal to the metric factor $g_{\boldsymbol{\xi}}$ (with $\boldsymbol{\xi}^{-1}$ similar to $\boldsymbol{g_\xi}$), this density matrix approach avoids solving Liouville's equation to obtain a metric factor compatible with the flow. Instead, one can compute the matrix $\boldsymbol{\xi}$ itself to determine the factor over time from numerical simulations of Lyapunov vectors.

B. Generalization of Liouville's equation

Several results follow from our identification of the relationship between the density matrix and the metric tensor. Most immediate is that Eq. 7 becomes the transformation of the metric determinant: $\sqrt{g_{\boldsymbol{\xi}}(t)}d\mathcal{V}(t) = \sqrt{g_{\boldsymbol{\xi}}(t_0)}d\mathcal{V}(t_0)$ with $g_{\boldsymbol{\xi}} = |\boldsymbol{\xi}^{-1}|$. This is a general form of Liouville's theorem, which is general in the sense that it is valid for non-Hamiltonian systems, relating the metric determinant [38, 42] to a geometric property of phase space.

What traditionally follows from Liouville's theorem is Liouville's equation, a formally exact equation of motion for the probability density in phase space [1]. This equation derives from another statement of Liouville's theorem: the density of representative points in the phase space is conserved along the trajectories of Hamiltonian systems, $d_t\rho(\boldsymbol{x}) = 0$ [2]. By contrast, we have defined a classical density matrix – a function of mechanical variables – in terms of Lyapunov vectors that describes the time evolution of phase space volume, not the statistical density [35]. Nevertheless, the classical density matrix gives a generalization of Liouville's equation as we show in Appendix B.

With the similarity of the unnormalized density matrix and the metric tensor, the flow compressibility accounts for the metric's compatibility with the dynamics [42], $-d_t \ln\sqrt{g_{\boldsymbol{\xi}}} = (n/2)d_t \ln \operatorname{Tr} \boldsymbol{g}_{\boldsymbol{\xi}}^{-1} = \boldsymbol{\nabla}\cdot\dot{\boldsymbol{x}}$. By identifying the density matrix as similar to the dual metric tensor, $\boldsymbol{g}_{\boldsymbol{\xi}}^{-1}$, we can also find this compatibility condition as the equation of motion for the metric determinant, Eq. 5, and $\operatorname{Tr}\boldsymbol{A}_+$. Therefore, $|\boldsymbol{\xi}|^{-\frac{1}{2}}$ obeys the generalized Liouville's equation:

$$\frac{\partial}{\partial t}|\boldsymbol{\xi}|^{-\frac{1}{2}} + \boldsymbol{\nabla}\cdot(|\boldsymbol{\xi}|^{-\frac{1}{2}}\dot{\boldsymbol{x}}) = 0, \tag{8}$$

for the evolution of the classical density matrix $|\boldsymbol{\xi}|$, defined in terms of the perturbations of state space variables.

While this Liouville equation applies to non-Hamiltonian systems, the equation of motion reduces to a simpler form if the dynamics are Hamiltonian. For Hamiltonian dynamics, the metric determinant $|\boldsymbol{\xi}|^{-1}$ is time-independent and the divergence of the flow vanishes, $d\mathcal{V}(t) = d\mathcal{V}(t_0)$ from Eq. 7. We also find,

$$\frac{\partial}{\partial t}|\boldsymbol{\xi}|^{-\frac{1}{2}} + \dot{\boldsymbol{x}}\cdot\boldsymbol{\nabla}|\boldsymbol{\xi}|^{-\frac{1}{2}} = 0, \tag{9}$$

an equation that is similar to the well-known form of the Liouville equation, with statistical density replaced with the determinant of $\boldsymbol{\xi}$.

The metric tensor, $\boldsymbol{g_\xi}$, and the density matrix, $\boldsymbol{\xi}^{-1}$, are related by a similarity transformation, so their determinants are equal. The determinant $|\boldsymbol{\xi}|$ is a mechanical function because the matrix $\boldsymbol{\xi}$ is determined by linearly independent tangent vectors forming a complete basis set at a given phase space point. It is the phase space volume squared $|\boldsymbol{\xi}| = (\delta q\delta p)^2$ for two-dimensional Hamiltonian systems described by $\boldsymbol{\xi} = \delta q\delta p\,\mathbb{1}_{2\times2}$, where $\mathbb{1}_{n\times n}$ is the $n\times n$ identity matrix. The basis tangent vectors define a locally conserved phase space volume at each of the phase space point. We discuss an important set of basis vectors for Hamiltonian systems in Sec. III B. As the density matrix $\boldsymbol{\xi}$ can be constructed locally using the tangent vectors, it makes the metric tensor numerically computable (up to a similarity transformation) and avoids having to solve the generalized version of the Liouville equation [42]; preserving time-reversibility of the Liouville equation can be a challenge in numerical solutions [43].

C. Poisson brackets

While we have focused on the determinant of the unnormalized density matrix, its trace also appears in the compatibility condition, Appendix Eq. B18. Analyzing the trace, we also find connections to classical dynamics, well-known quantities in dynamical systems. Because of their use in quantum-mechanical expectation values, one might expect traces to also quantify useful observables in this classical setting. Here, the dynamics of $\boldsymbol{\xi} = \sum_{i=1}^{k}|\delta\boldsymbol{\psi}_i\rangle\langle\delta\boldsymbol{\psi}_i|$ are not trace preserving, but if $\boldsymbol{\xi}$ is built from a perturbation to a phase point, its trace yields the magnitude of the perturbation. So, the trace over $\boldsymbol{\xi}$ does give us a new perspective on a well-known quantity with physical implications, Lyapunov exponents.

The rate of change of $\operatorname{Tr}\boldsymbol{\xi}$,

$$\frac{1}{2}\frac{d}{dt}\operatorname{Tr}\boldsymbol{\xi} = \frac{1}{2}\operatorname{Tr}\{\boldsymbol{A}_+,\boldsymbol{\xi}\} = \operatorname{Tr}(\boldsymbol{\xi}\boldsymbol{A}_+) = \langle\boldsymbol{A}_+\rangle_{\boldsymbol{\xi}}, \tag{10}$$

is set by $\langle\boldsymbol{A}_+\rangle_{\boldsymbol{\xi}_i}$, a quantity related to the instantaneous Lyapunov exponents [21], which are a measure of local (in)stability. We show in App. A that $\operatorname{Tr}(\boldsymbol{\xi}\boldsymbol{A}_+) = \langle\boldsymbol{A}_+\rangle_{\boldsymbol{\xi}}$.

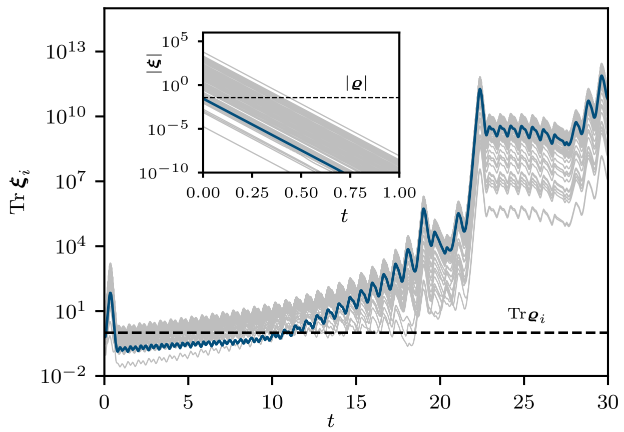

FIG. 2. For the Lorenz-Fetter model, the trace of the unnormalized density matrix, $\boldsymbol{\xi}_i(t)$ (see Eq. 2 for $k = 1$), as a function of time for 100 tangent vectors. Each vector is a local perturbation with elements sampled from a uniform distribution. The inset shows the evolution of $|\boldsymbol{\xi}_i(t)|$. For each tangent vector, we express $\boldsymbol{\xi}_i$ in terms of its complete set of linearly independent basis vectors. The determinant in each case decays at rate given by instantaneous Lyapunov exponent $\langle \boldsymbol{A}_+ \rangle$. An example of a normalized tangent vector evolving on the Lorenz attractor in Fig. 1 is shown in blue. The trace and determinant of the normalized density matrix $\boldsymbol{\varrho}$ (inset) are time-invariant (dashed).

Defining $\langle \boldsymbol{A}_+ \rangle = \langle \boldsymbol{A}_+ \rangle_{\boldsymbol{\xi}} / \operatorname{Tr} \boldsymbol{\xi}$, the solution to this equation of motion is:

$$\operatorname{Tr} \boldsymbol{\xi}(t) = \operatorname{Tr} \boldsymbol{\xi}(t_0) e^{2 \int_{t_0}^{t} \langle \boldsymbol{A}_+(t') \rangle \, dt'}. \tag{11}$$

(In the next section, we identify the quantity $\langle \boldsymbol{A}_+ \rangle$ as the instantaneous Lyapunov exponent.)

To numerically verify this result, and others, we simulated the dynamics of Hamiltonian and dissipative dynamical systems. As a prototypical dissipative system, we chose the Lorenz-Fetter model. Figure 2 shows the time evolution of the trace for a chaotic orbit for 100 random perturbation states drawn from a uniform distribution. The rapid increase in $\operatorname{Tr} \boldsymbol{\xi}_i$ (on the semi-log scale) is indicative of the chaotic nature of the chosen orbit.

For Hamiltonian systems, the equation of motion for the trace can be expressed as a Poisson bracket. In classical statistical mechanics, a dynamical variable, $f(\boldsymbol{q}, \boldsymbol{p})$, can be expressed as $\dot{f} = \{f, H\}_P$, in terms of the Poisson bracket, $\{.\}_P$. Combined with our result above, this fact gives a correspondence,

$$\operatorname{Tr} \dot{\boldsymbol{\xi}} = 2 \langle \boldsymbol{\mathcal{H}}_+ \rangle_{\boldsymbol{\xi}} = \{\operatorname{Tr} \boldsymbol{\xi}, H\}_P, \tag{12}$$

between the Poisson bracket and an average $\langle \boldsymbol{\mathcal{H}}_+ \rangle_{\boldsymbol{\xi}}$ of the symmetric part of the stability matrix $\boldsymbol{\mathcal{H}}_+$. For example, for an arbitrary perturbation $(\delta q, \delta p)^\top)$, in the phase space of the linear harmonic oscillator, $\operatorname{Tr} \boldsymbol{\xi} = \delta q^2 + \delta p^2$ (where $\boldsymbol{\xi}$ here is from Eq. 2 with $k = 1$) and $\operatorname{Tr} \dot{\boldsymbol{\xi}} = \{\operatorname{Tr} \boldsymbol{\xi}, H\}_P = 2 \delta q \delta p (1 - \omega^2)$ where ω is the oscillation frequency. As another example, Figure 3(a) shows $\operatorname{Tr} \boldsymbol{\xi}$ for the classical Hénon-Heiles system on a regular and chaotic orbit (with $\boldsymbol{\xi}$ built from a single perturbation vector.

III. DYNAMICS OF THE CLASSICAL NORMALIZED DENSITY MATRIX

So far, we have shown that the density matrix is similar to the phase space metric, thus defining the underlying geometry, with its determinant being the phase space volume element. However, the key results differ in some respects from their analogues in quantum mechanics. For example, the dynamics of $\boldsymbol{\xi}$ are not norm-preserving, despite the norm-preserving dynamics of Hilbert state vectors being a basic postulate of many formulations of quantum mechanics. To sharpen the correspondence with quantum mechanics, we can derive a norm-preserving dynamics for the classical density matrix. These dynamics for the normalized density matrix (and its properties) have implications for classical statistical physics. In particular, this normalization permits the definition of "averages" that define observables, such as instantaneous Lyapunov exponents.

In order to derive the dynamics of general, classical systems, we consider a unit tangent vector to the state-space variables $|\delta \boldsymbol{u}\rangle = |\delta \boldsymbol{x}\rangle / \|\delta \boldsymbol{x}\|$, where $\|.\|$ is the ℓ_2-norm. The vector has the equation of motion,

$$\frac{d}{dt} |\delta \boldsymbol{u}\rangle = (\boldsymbol{A}_+ + \boldsymbol{A}_-) \, |\delta \boldsymbol{u}\rangle - r \, |\delta \boldsymbol{u}\rangle \,, \tag{13}$$

contains a source/sink term with the instantaneous rate: $r := r(t) = \langle \delta \boldsymbol{u} | \, \boldsymbol{A}_+ \, | \delta \boldsymbol{u} \rangle = d_t \ln \|\delta \boldsymbol{x}(t)\|$. This rate is the instantaneous Lyapunov exponent (or local stretching rate) for a linearized dynamics, which is related to the finite-time Lyapunov exponent,

$$\lambda(t) := \lambda(t, t_0) = |t - t_0|^{-1} \int_{t_0}^{t} r(t) \, dt. \tag{14}$$

The maximum instantaneous Lyapunov exponent is also referred to as *reactivity* – the maximum amplification rate over all perturbations, immediately following a perturbation [44]. Even in asymptotically stable systems with this instability can exhibit transient behavior [45] in response to external stimuli [46]. In the long time limit, the time average of this expansion rate is the Lyapunov exponent, $\lambda = \lim_{t \to \infty} \lambda_i(t)$, which is independent of initial conditions [47].

As before, we represent the state of the dynamical system $\dot{\boldsymbol{x}} = \boldsymbol{F}$ as a density matrix. But now, we express it in terms of a unit tangent-space basis $\{|\delta \boldsymbol{\phi}_i\rangle\}$. Normalizing each $|\delta \boldsymbol{\phi}_i\rangle = c_i \, |\delta \boldsymbol{\psi}_i\rangle$, we can define the pure states with the expected properties: $\boldsymbol{\varrho}_i^2 = \boldsymbol{\varrho}_i$, $\operatorname{Tr} \boldsymbol{\varrho}_i = 1$, $\operatorname{Tr} \boldsymbol{\varrho}_i^2 = 1$, symmetric, $\boldsymbol{\varrho}_i \succeq 0$, i.e., $\boldsymbol{\varrho}_i$ is positive semi-definite. Proving these properties requires the dynamics of $\boldsymbol{\varrho}_i$ be norm-preserving. We refer to this *normalized*

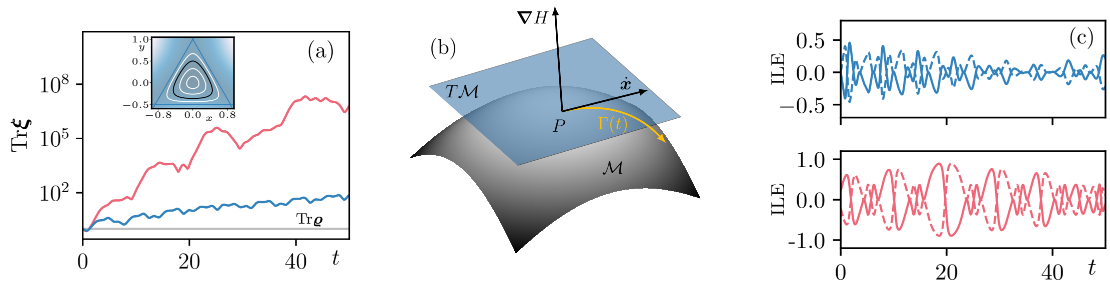

FIG. 3. (a) The trace of the unnormalized density matrices for a regular (blue) and a chaotic orbit (red) for the Hénon-Heiles system. The trace of the normalized density matrices is time invariant (gray). The inset shows the equipotential surface (and lines) for the potential $V = \frac{1}{2}(x^2 + y^2 + 2x^2y - \frac{2}{3}y^3)$. Line colors in (a) correspond to those of the equipotential lines. (b) Schematic illustration of the neighborhood of a point P along a trajectory $\Gamma(t)$ through the phase space $\mathcal{M}$ with the associated tangent space $\mathcal{TM}$ and conjugate vectors, $\boldsymbol{\nabla}H$ and $\dot{\boldsymbol{x}}$. (c) Pair of instantaneous Lyapunov exponents (ILE) for the regular with $E = 0.0833$ (blue) and a chaotic orbit with $E = 0.1667$ (red).

density matrix $\boldsymbol{\varrho}$ as a "perturbation state":

$$\boldsymbol{\varrho}(t) = \frac{\boldsymbol{\xi}(t)}{\operatorname{Tr}\boldsymbol{\xi}(t)} = \mathcal{C}^{-1}\sum_{i=1}^{k} c_i^2 \,|\delta\boldsymbol{\psi}_i\rangle\langle\delta\boldsymbol{\psi}_i|\,, \tag{15}$$

which is directly related to the unnormalized $\boldsymbol{\xi}$ with $\operatorname{Tr}\boldsymbol{\xi} = \sum_{i=1}^{k} c_i^2 = \mathcal{C}$ so that $\operatorname{Tr}\boldsymbol{\varrho} = 1$. The tangent-space average $\langle\delta\boldsymbol{\phi}|\boldsymbol{\varrho}|\delta\boldsymbol{\phi}\rangle$ over the unit basis is analogous to the quantum-mechanical probability of finding the system at $|\delta\boldsymbol{\phi}\rangle$ given that its state is $\boldsymbol{\varrho}$; for example, if all tangent space directions contribute equally $\langle\delta\boldsymbol{\phi}|\boldsymbol{\varrho}|\delta\boldsymbol{\phi}\rangle = k^{-1}$.

With these normalized states, we can define tangent space averages that are physical observables. For instance, the entropy production for thermostatted systems [36] is related to the phase space contraction rate, which derives directly from the density matrix. Consider a maximally-mixed state $\boldsymbol{\varrho}^M$ for $k = n$ and n phase space dimensions. If the state is maximally mixed [4] with $c_i = 1\ \forall i$, then $\operatorname{Tr}\boldsymbol{\xi} = k$. The phase space volume contraction rate can be expressed as a tangent-space average of $\boldsymbol{A}_+$ over $\boldsymbol{\varrho}$:

$$\Lambda = \langle\boldsymbol{A}_+\rangle_{\boldsymbol{\varrho}^M} = n^{-1}\operatorname{Tr}\big(\boldsymbol{A}_+\boldsymbol{\varrho}^M\big) = n^{-1}\sum_{i=1}^{n}\operatorname{Tr}(\boldsymbol{A}_+\boldsymbol{\varrho}_i),$$

where each of the instantaneous Lyapunov exponents is also a tangent space average of $\boldsymbol{A}_+$ over pure *basis* states $\boldsymbol{\varrho}_i$: $r_i = \langle\boldsymbol{A}\rangle_{\boldsymbol{\varrho}_i} = \operatorname{Tr}(\boldsymbol{A}_+\boldsymbol{\varrho}_i)$.

The normalized state (pure or maximally mixed) evolves in time according to:

$$\frac{d\boldsymbol{\varrho}}{dt} = \{\boldsymbol{A}_+, \boldsymbol{\varrho}\} + [\boldsymbol{A}_-, \boldsymbol{\varrho}] - 2r\boldsymbol{\varrho}, \tag{16}$$

another classical analogue of the von Neumann equation in quantum mechanics. From this equation, we see that the average of $\boldsymbol{A}_+$ at each instant of time, i.e., the instantaneous Lyapunov exponent, is crucial to norm preservation. It offsets the stretching and contraction of $|\delta\boldsymbol{u}\rangle$ due to $\boldsymbol{A}$ along a given trajectory. Solving this equation of motion, we find the density matrix $\boldsymbol{\varrho}$ at time t_0 and at a later time t are similar:

$$\boldsymbol{\varrho}(t) = \tilde{\boldsymbol{M}}(t,t_0)\boldsymbol{\varrho}(t_0)\tilde{\boldsymbol{M}}^{-1}(t,t_0), \tag{17}$$

when $\boldsymbol{\varrho}$ is composed of vectors $\{|\delta\boldsymbol{u}^i\rangle\}$ evolving in time under $\tilde{\boldsymbol{M}}$. Regardless of the underlying dynamics, we can define the norm-preserving evolution matrix $\tilde{\boldsymbol{M}} := \boldsymbol{M}\boldsymbol{\Gamma}$. It is generally non-orthogonal with $|\tilde{\boldsymbol{M}}| = 1$, making it a unimodular matrix that generates volume-preserving transformations, App. A. The matrix $\boldsymbol{\Gamma}$ has the inverse expansion factors $\|\delta\boldsymbol{x}^i(t_0)\|/\|\delta\boldsymbol{x}^i(t)\| = e^{-\lambda_i(t)(t-t_0)}$ on the diagonal.

A. Generalized Liouville's theorem/equation

Imposing a norm-preserving dynamics not only normalizes the density matrix, it also reinstates the original form of Liouville's equation for non-Hamiltonian dynamics. To see this result, we again take the basis to span the phase space $k \to n$, i.e., a complete basis. As must be the case for a norm-preserving dynamics, the magnitude of perturbations do not evolve with time and $\operatorname{Tr}\boldsymbol{\varrho}(t) = \operatorname{Tr}\boldsymbol{\varrho}(t_0) = 1$. The preservation of the trace $\operatorname{Tr}\boldsymbol{\varrho}$ follows from Eq. 16, which shows $\operatorname{Tr}\dot{\boldsymbol{\varrho}} = 0$ because $\operatorname{Tr}\{\boldsymbol{A}_+, \boldsymbol{\varrho}\} = 2r$. However, both the trace and the determinant

$$|\boldsymbol{\varrho}(t)| = |\boldsymbol{\varrho}(t_0)|, \tag{18}$$

are similarity-invariant constants of motion, Figs. 2 and 3. Defining the metric tensor $\boldsymbol{g}_{\boldsymbol{\varrho}}$ such that its determinant $g_{\boldsymbol{\varrho}} = |\boldsymbol{\varrho}^{-1}|$, the conservation of the determinant is another form of the Liouville's equation for non-Hamiltonian systems,

$$\frac{\partial}{\partial t}|\boldsymbol{\varrho}|^{-\frac{1}{2}} + \dot{\boldsymbol{x}}\cdot\boldsymbol{\nabla}|\boldsymbol{\varrho}|^{-\frac{1}{2}} = 0. \tag{19}$$

in terms of $\boldsymbol{\varrho}$. The compatibility condition of the metric is $\partial_t\sqrt{g_{\boldsymbol{\varrho}}} + \dot{\boldsymbol{x}}\cdot\boldsymbol{\nabla}\sqrt{g_{\boldsymbol{\varrho}}} = 0$. For Hamiltonian dynamics, this condition has the same form as that of $g_{\boldsymbol{\xi}}$, App. B, and the well-known Liouville equation for statistical density. However, this generalization using the normalized matrix $\boldsymbol{\varrho}$ (and the compatibility condition for $g_{\boldsymbol{\varrho}}$) is valid regardless of whether the dynamics are Hamiltonian or not and has a form that is similar to the usual Liouville's equation.

Imposing a norm-preserving dynamics and normalizing the density matrix also reinstates the form of the traditional Liouville theorem for non-Hamiltonian dynamics. With these norm-preserving dynamics, the generalized Liouville theorem in Eq. 7 becomes: $|\boldsymbol{\Gamma}|d\mathcal{V}(t) = |\boldsymbol{M}||\boldsymbol{\Gamma}|d\mathcal{V}(t_0) = |\tilde{\boldsymbol{M}}|d\mathcal{V}(t_0) = d\mathcal{V}(t_0)$ and we can recognize $|\boldsymbol{\Gamma}|^2 = |\boldsymbol{\xi}(t_0)|/|\boldsymbol{\xi}(t)|$. Geometrically, the generalized Liouville theorem here is: scaled phase space volumes are conserved under the norm-preserving evolution of a perturbation state with a basis that spans the n-dimensional phase space. This generalization of Liouville's theorem is not limited to Hamiltonian dynamics, however. In the phase space of non-Hamiltonian systems, any part of the initial volume lost (or gained) in course of the dynamics is continually and entirely compensated for by the stretching/contracting of the volume. As a result, the scaled volume $|\boldsymbol{\Gamma}|d\mathcal{V}$ is conserved for the n-dimensional phase space of any dynamical system.

B. Basis representation

In quantum statistical mechanics, the choice of basis states provides an explicit matrix representation of the quantum state. Here, there are also natural sets of vectors one might choose for the classical state in the tangent space: $\boldsymbol{\varrho}$ and $\boldsymbol{\xi}$. In dynamical systems theory, it is common to analyze Lyapunov vectors, such as Gram-Schmidt vectors [48, 49]. More recently, however, there has been an interest in covariant Lyapunov vectors [29]. Based on early work [50], Lyapunov vectors with small, but finite, exponents are hydrodynamic modes that characterize macroscopic transport [51, 52]. Any of these sets of vectors can be used to construct classical density matrices.

There are other choices for basis vectors that are natural to the nonlinear dynamics and give another connection to Liouville's theorem. For example, Hamiltonian systems have special tangent space directions associated with conserved quantities. On a constant energy manifold, for example, there are two conjugate tangent directions: the phase velocity $\dot{\boldsymbol{x}}$ and the gradient of the Hamiltonian $\boldsymbol{\nabla}H$, Fig. 3(b). They are related as $\dot{\boldsymbol{x}} = \boldsymbol{\Omega}\boldsymbol{\nabla}H$ through the Poisson matrix $\boldsymbol{\Omega}$, orthogonal to each other, $\boldsymbol{\nabla}H\cdot\boldsymbol{\Omega}\boldsymbol{\nabla}H = 0$, and have equal magnitude $\|\dot{\boldsymbol{x}}\| = \|\boldsymbol{\nabla}H\|$ through Hamilton's equations. The vector $\boldsymbol{\nabla}H$ is also orthogonal to the constant energy manifold and used to define the invariant measure [9]. In general, the vector sets defining the density matrices here need not span the whole phase space. So, forming density matrices from each conjugate vector, we can find the pure states, $\boldsymbol{\varrho}_{\dot{\boldsymbol{x}}}$ and $\boldsymbol{\varrho}_{\boldsymbol{\Omega}\boldsymbol{\nabla}H}$. These conjugate pure states in the tangent space for a two-dimensional Hamiltonian system are:

$$\begin{aligned} \|\dot{\boldsymbol{x}}\|^2\boldsymbol{\varrho}_{\dot{\boldsymbol{x}}} &= \begin{pmatrix} \dot{q}^2 & \dot{p}\dot{q} \\ \dot{p}\dot{q} & \dot{p}^2 \end{pmatrix}, \\ \|\boldsymbol{\nabla}H\|^2\boldsymbol{\varrho}_{\boldsymbol{\nabla}H} &= \begin{pmatrix} \dot{p}^2 & -\dot{p}\dot{q} \\ -\dot{p}\dot{q} & \dot{q}^2 \end{pmatrix}. \end{aligned} \tag{20}$$

These states are formed from the outer product of the unit tangent vectors, $|\delta\boldsymbol{\phi}_{\dot{\boldsymbol{x}}}\rangle = \|\dot{\boldsymbol{x}}\|^{-1}(\dot{q},\dot{p})^\top$ and $|\delta\boldsymbol{\phi}_{\boldsymbol{\nabla}H}\rangle = \|\boldsymbol{\nabla}H\|^{-1}(-\dot{p},\dot{q})^\top$, where $\|\dot{\boldsymbol{x}}\|^2 = \|\boldsymbol{\nabla}H\|^2 = \dot{p}^2 + \dot{q}^2$. Both density matrices have unit trace and are related by $\boldsymbol{\varrho}_{\dot{\boldsymbol{x}}} = \boldsymbol{\Omega}\boldsymbol{\varrho}_{\boldsymbol{\nabla}H}\boldsymbol{\Omega}^\top$.

These particular density matrices also have a (lower dimensional) parallel with Liouville's theorem. Liouville's theorem can be thought of as an equivalence of the divergence of the phase flow, the trace of the Jacobian, and the intrinsic rate: $d_t \ln\delta\mathcal{V} = \boldsymbol{\nabla}\cdot\dot{\boldsymbol{x}} = n\,\mathrm{Tr}[\boldsymbol{\varrho}\boldsymbol{\mathcal{H}}(\boldsymbol{x})]$, for $\boldsymbol{\varrho} = n^{-1}\sum_{i=1}^n |\delta\boldsymbol{\phi}_i\rangle\langle\delta\boldsymbol{\phi}_i|$. There is a similar equivalence for the instantaneous Lyapunov exponents of $\dot{\boldsymbol{x}}$ and $\boldsymbol{\nabla}H$. For a two-dimensional Hamiltonian system, these take the form:

$$\begin{aligned} \frac{d\ln\|\dot{\boldsymbol{x}}\|}{dt} &= \frac{\dot{q}\cdot\dot{p}}{\|\dot{\boldsymbol{x}}\|^2}\boldsymbol{\nabla}\cdot\begin{pmatrix}\dot{p}\\ \dot{q}\end{pmatrix} = \mathrm{Tr}(\boldsymbol{\varrho}_{\dot{x}}\boldsymbol{\mathcal{H}}_+), \\ \frac{d\ln\|\boldsymbol{\nabla}H\|}{dt} &= \frac{-\dot{q}\cdot\dot{p}}{\|\boldsymbol{\nabla}H\|^2}\boldsymbol{\nabla}\cdot\begin{pmatrix}\dot{p}\\ \dot{q}\end{pmatrix} = \mathrm{Tr}(\boldsymbol{\varrho}_{\boldsymbol{\nabla}H}\boldsymbol{\mathcal{H}}_+), \end{aligned} \tag{21}$$

denoting the phase point $\boldsymbol{x} = (q,p)$. Unlike, the intrinsic rate of the volume element $\delta\mathcal{V} = \delta q\delta p$ that appears in Liouville's theorem, these instantaneous Lyapunov exponents are not zero – they are related to the divergence in a common direction $(\dot{p},\dot{q})^\top$ that is a reflection about $\dot{p} = \dot{q}$. Instead, they are conjugate, so they sum to zero. To illustrate, we show the instantaneous Lyapunov exponents in the tangent space directions computed from Eq. 21 for the Hénon-Heiles systems in Fig. 3(c). A vanishing sum of instantaneous Lyapunov exponents corresponds to conservation of phase space volume.

IV. CONCLUSIONS

Liouville's equation and theorem are the foundation of statistical mechanics established by Gibbs, Maxwell, and Boltzmann. Boltzmann, for example, approximated Liouville's equation to derive his H-theorem for irreversible processes, making an assumption of "molecular chaos". Here, we have established a density matrix formulation of dynamical systems that explicitly and quantitatively accounts for measures of local instability and chaos, Lyapunov exponents, and the phase space contraction rate associated with dissipation. The classical density matrix defines a phase space geometry of the deterministic dynamics and gives a means of computing statistical mechanical observables such as energy dissipation. Through this connection, we could derive generalizations Liouville's theorem/equation for any differentiable dynamical system. Many dynamical systems are not Hamiltonian, including those that are dissipative, non-steady, and driven, for which the Liouville's equation/theorem here still apply. And, when the dynamics are Hamiltonian, these generalizations reduce to the traditional forms of the Liouville theorem and Liouville's equation. We have shown they derive from the properties of classical density matrices, which themselves evolve under an equation of motion akin to the von Neumann equation at the foundation of quantum statistical mechanics. From these results, the generalized Liouville equation becomes numerically computable and, thus, a new basis for analyzing classical speed limits on observables [53], the spread of perturbations, and the transport of statistical density in dynamical systems.

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Appendix A: Equation of motion for Lyapunov vectors

Consider the time evolution equation of a generic, infinitesimal perturbation $|\delta\boldsymbol{x}\rangle \in T\mathcal{M}$,

$$\frac{d}{dt}\left|\delta\boldsymbol{x}(t)\right\rangle = \boldsymbol{A}(\boldsymbol{x})\left|\delta\boldsymbol{x}(t)\right\rangle, \tag{A1}$$

governed by the stability matrix of the system, $\boldsymbol{A}$. Using the time-ordering operator $\mathcal{T}_+$, perturbations propagate as:

$$\begin{aligned}\left|\delta\boldsymbol{x}(t)\right\rangle &= \boldsymbol{M}(t,t_0)\left|\delta\boldsymbol{x}(t_0)\right\rangle \\ &= \mathcal{T}_+ e^{\int_{t_0}^{t}\boldsymbol{A}(t')\,dt'}\left|\delta\boldsymbol{x}(t_0)\right\rangle.\end{aligned} \tag{A2}$$

The evolution operator or Jacobian matrix $[\boldsymbol{M}(t,t_0)]^i_{\ j} = \partial x^i(t)/\partial x^j(t_0)$ has the equation of motion:

$$\frac{d\boldsymbol{M}}{dt} = \boldsymbol{A}\boldsymbol{M} \qquad \text{or} \qquad \boldsymbol{A} = \frac{d\boldsymbol{M}}{dt}\boldsymbol{M}^{-1}. \tag{A3}$$

The determinant obeys Jacobi's formula:

$$\frac{d}{dt}|\boldsymbol{M}| = |\boldsymbol{M}|\,\mathrm{Tr}\left(\frac{d\boldsymbol{M}}{dt}\boldsymbol{M}^{-1}\right) = |\boldsymbol{M}|\,\mathrm{Tr}\,\boldsymbol{A}. \tag{A4}$$

Each $|\delta\boldsymbol{x}\rangle \in T\mathcal{M}$ has a corresponding $\langle\delta\boldsymbol{x}| \in T\mathcal{M}$. To find the dynamics of the dual vector $\langle\delta\dot{\boldsymbol{x}}(t)|$, we partition the stability matrix $\boldsymbol{A} = \boldsymbol{A}_+ + \boldsymbol{A}_-$ into its symmetric and anti-symmetric parts, $\boldsymbol{A}_\pm = \frac{1}{2}(\boldsymbol{A} \pm \boldsymbol{A}^\top)$. Dual vectors then evolve according to:

$$\frac{d}{dt}\left\langle\delta\boldsymbol{x}(t)\right| = \left\langle\delta\boldsymbol{x}(t)\right|(\boldsymbol{A}_+ - \boldsymbol{A}_-). \tag{A5}$$

Together, the equations for the motion of tangent vectors and their dual define the non-unitary dynamics of $|\delta\boldsymbol{x}\rangle$ in the tangent space, $\langle\delta\boldsymbol{x}(t)|\delta\boldsymbol{x}(t)\rangle \neq \langle\delta\boldsymbol{x}(t_0)|\delta\boldsymbol{x}(t_0)\rangle$.

The time evolution of a unit Lyapunov vector $|\delta\boldsymbol{u}\rangle$ in the phase space of a dynamics system,

$$\frac{d}{dt}\left|\delta\boldsymbol{u}\right\rangle = \boldsymbol{A}_+\left|\delta\boldsymbol{u}\right\rangle + \boldsymbol{A}_-\left|\delta\boldsymbol{u}\right\rangle - r\left|\delta\boldsymbol{u}\right\rangle, \tag{A6}$$

has an additional source/sink term with the instantaneous Lyapunov exponent $r = \langle\boldsymbol{A}_+\rangle = \langle\delta\boldsymbol{u}|\,\boldsymbol{A}_+\,|\delta\boldsymbol{u}\rangle$. The solution is:

$$\left|\delta\boldsymbol{u}\right\rangle = (\boldsymbol{M}\boldsymbol{\Gamma})\left|\delta\boldsymbol{u}(t_0)\right\rangle =: \tilde{\boldsymbol{M}}(t,t_0)\left|\delta\boldsymbol{u}(t_0)\right\rangle. \tag{A7}$$

For any dynamics through the state space, the norm-preserving evolution operator $\tilde{\boldsymbol{M}} := \boldsymbol{M}\boldsymbol{\Gamma}$ is not necessarily orthogonal but $|\tilde{\boldsymbol{M}}| = 1$. The matrix $\boldsymbol{\Gamma}$ has the inverse expansion factors $\|\delta\boldsymbol{x}_i(t_0)\|/\|\delta\boldsymbol{x}_i(t)\| = e^{-\lambda_i(t)(t-t_0)}$ on the diagonal. The equation of motion for $\langle\delta\boldsymbol{u}|$,

$$\frac{d}{dt}\left\langle\delta\boldsymbol{u}\right| = \left\langle\delta\boldsymbol{u}\right|\boldsymbol{A}_+ - \left\langle\delta\boldsymbol{u}\right|\boldsymbol{A}_- - \left\langle\boldsymbol{A}_+\right\rangle\left\langle\delta\boldsymbol{u}\right|, \tag{A8}$$

has the solution:

$$\left\langle\delta\boldsymbol{u}\right| = \left\langle\delta\boldsymbol{u}(t_0)\right|(\boldsymbol{M}\boldsymbol{\Gamma})^\top =: \left\langle\delta\boldsymbol{u}(t_0)\right|\tilde{\boldsymbol{M}}^\top(t,t_0). \tag{A9}$$

The equation of motion for density matrices follow from these results for tangent vectors. In the main text, we consider basis sets that span the n-dimensional phase space, defining the unnormalized,

$$\boldsymbol{\xi} = \sum_{i=1}^{k}\left|\delta\boldsymbol{\psi}_i\right\rangle\!\left\langle\delta\boldsymbol{\psi}_i\right| = \sum_{i=1}^{k} c_i^2\left|\delta\boldsymbol{\phi}_i\right\rangle\!\left\langle\delta\boldsymbol{\phi}_i\right|, \tag{A10}$$

evolving as

$$\begin{aligned}\frac{d\boldsymbol{\xi}}{dt} &= \sum_{i=1}^{k}\left(\frac{d}{dt}\left|\delta\boldsymbol{\psi}_i\right\rangle\right)\left\langle\delta\boldsymbol{\psi}_i\right| + \left|\delta\boldsymbol{\psi}_i\right\rangle\left(\frac{d}{dt}\left\langle\delta\boldsymbol{\psi}_i\right|\right) \\ &= \sum_{i=1}^{k}\boldsymbol{A}_+\left|\delta\boldsymbol{\psi}_i(t)\right\rangle\!\left\langle\delta\boldsymbol{\psi}_i(t)\right| + \sum_{i=1}^{k}\boldsymbol{A}_-\left|\delta\boldsymbol{\psi}_i(t)\right\rangle\!\left\langle\delta\boldsymbol{\psi}_i(t)\right| \\ &\quad + \sum_{i=1}^{k}\left|\delta\boldsymbol{\psi}_i(t)\right\rangle\!\left\langle\delta\boldsymbol{\psi}_i(t)\right|\boldsymbol{A}_+ - \sum_{i=1}^{k}\left|\delta\boldsymbol{\psi}_i(t)\right\rangle\!\left\langle\delta\boldsymbol{\psi}_i(t)\right|\boldsymbol{A}_- \\ &= \boldsymbol{A}_+\boldsymbol{\xi} + \boldsymbol{A}_-\boldsymbol{\xi} + \boldsymbol{\xi}\boldsymbol{A}_+ - \boldsymbol{\xi}\boldsymbol{A}_- \\ &= \{\boldsymbol{A}_+, \boldsymbol{\xi}\} + [\boldsymbol{A}_-, \boldsymbol{\xi}],\end{aligned}$$

where we used Eq. A5 and its corresponding equation for $|\delta\boldsymbol{\psi}_i\rangle$.

Normalizing $\boldsymbol{\xi}$, we obtain the matrix:

$$\boldsymbol{\varrho}(t) = \mathcal{C}^{-1}\sum_{i=1}^{k} c_i^2\,\boldsymbol{\varrho}_i = \mathcal{C}^{-1}\sum_{i=1}^{k} c_i^2\,|\delta\boldsymbol{\phi}_i\rangle\langle\delta\boldsymbol{\phi}_i|\,. \tag{A11}$$

Here, $\mathcal{C} = \mathrm{Tr}\,\boldsymbol{\xi} = \sum_{i=1}^{k} c_i^2$. Each pure state $\boldsymbol{\varrho}_i$ evolves as:

$$\begin{aligned}\frac{d\boldsymbol{\varrho}_i}{dt} &= \frac{d}{dt}(|\delta\boldsymbol{\phi}_i\rangle\langle\delta\boldsymbol{\phi}_i|)\\ &= |\delta\boldsymbol{\phi}_i\rangle\left(\frac{d}{dt}\langle\delta\boldsymbol{\phi}_i|\right) + \left(\frac{d}{dt}|\delta\boldsymbol{\phi}_i\rangle\right)\langle\delta\boldsymbol{\phi}_i|\\ &= \boldsymbol{\varrho}_i\boldsymbol{A}_+ - \boldsymbol{\varrho}_i\boldsymbol{A}_- + \boldsymbol{A}_+\boldsymbol{\varrho}_i + \boldsymbol{A}_-\boldsymbol{\varrho}_i - 2r\,\boldsymbol{\varrho}_i\\ &= \{\boldsymbol{A}_+, \boldsymbol{\varrho}_i\} + [\boldsymbol{A}_-, \boldsymbol{\varrho}_i] - 2r_i\boldsymbol{\varrho}_i.\end{aligned} \tag{A12}$$

Appendix B: Generalized Liouville theorem and equation

For the unnormalized density matrix $\boldsymbol{\xi}$,

$$\ln|\boldsymbol{\xi}| = \mathrm{Tr}\ln\boldsymbol{\xi}, \tag{B1}$$

follows from the identity $\ln|\boldsymbol{C}| = \mathrm{Tr}\ln\boldsymbol{C}$ between the trace and the determinant $|.|$. Assuming $\boldsymbol{\xi}$ is invertible, taking the time derivative,

$$\frac{d}{dt}\ln|\boldsymbol{\xi}| = \mathrm{Tr}\,\frac{d}{dt}\ln\boldsymbol{\xi} = \mathrm{Tr}\,\boldsymbol{\xi}^{-1}\frac{d\boldsymbol{\xi}}{dt}, \tag{B2}$$

and using Eq. 3, we find the generalized Liouville's equation in the main text:

$$\frac{d}{dt}\ln|\boldsymbol{\xi}| = \mathrm{Tr}\,\boldsymbol{\xi}^{-1}(\{\boldsymbol{A}_+, \boldsymbol{\xi}\} + [\boldsymbol{A}_-, \boldsymbol{\xi}]) = 2\,\mathrm{Tr}\,\boldsymbol{A}_+. \tag{B3}$$

To see that this result is a generalization of Liouville's equation requires recognizing the phase space volume element is $d\mathcal{V} = dx^1 \wedge \cdots \wedge dx^n$. The determinant of the Jacobian $\boldsymbol{M}$ governs its coordinate transformation under the action of the dynamical equations: $d\mathcal{V}(t) = |\boldsymbol{M}(t,t_0)|d\mathcal{V}(t_0)$. Equation 7 follows from the determinant of Eq. 4: $|\boldsymbol{\xi}(t)| = |\boldsymbol{M}\,(t,t_0)\,|^2|\boldsymbol{\xi}\,(t_0)\,|$.

Treating the phase space as a general Riemannian manifold endowed with a (contravariant) metric tensor $\boldsymbol{g}_{\boldsymbol{\xi}}^{-1}$, we identify this metric tensor as *similar* to the unnormalized density matrix $\boldsymbol{\xi}$. To see this relationship, consider two arbitrary ordered bases $|\delta\boldsymbol{\psi}_i(t_0)\rangle$ and $|\delta\boldsymbol{\psi}_i(t)\rangle$ stacked in matrix columns,

$$\boldsymbol{\Psi'} := \boldsymbol{\Psi}(t_0) = \Big[\,|\delta\boldsymbol{\psi}_1(t_0)\rangle\,,\cdots,|\delta\boldsymbol{\psi}_n(t_0)\rangle\,\Big] \tag{B4}$$

$$\boldsymbol{\Psi}(t) = \Big[\,|\delta\boldsymbol{\psi}_1(t)\rangle\,,\cdots,|\delta\boldsymbol{\psi}_n(t)\rangle\,\Big], \tag{B5}$$

and related by $\boldsymbol{\Psi'} = \boldsymbol{M}\boldsymbol{\Psi}$. In these bases, we can represent the transformation of the unnormalized density matrix:

$$\boldsymbol{\xi}(t) = \boldsymbol{\Psi'}\boldsymbol{\Psi'}^{\top} = \boldsymbol{M}\boldsymbol{\Psi}\boldsymbol{\Psi}^{\top}\boldsymbol{M}^{\top} = \boldsymbol{M}\boldsymbol{\xi}(t_0)\boldsymbol{M}^{\top}. \tag{B6}$$

However, the linear transformation $\boldsymbol{\Psi} \to \boldsymbol{\Psi'}$ is also obtained by a change-of-basis matrix $\boldsymbol{P}$ as $\boldsymbol{\Psi'} = \boldsymbol{\Psi}\boldsymbol{P}$. The Jacobian $\boldsymbol{M}$ and $\boldsymbol{P}$ are related by a similarity transformation

$$\boldsymbol{P} = \boldsymbol{\Psi}^{-1}\boldsymbol{M}\boldsymbol{\Psi}. \tag{B7}$$

One can then view $\boldsymbol{P}$ and $\boldsymbol{M}$ as *propagators* expressed in different bases that represent the linear transformation of $\boldsymbol{\Psi} \to \boldsymbol{\Psi'}$ forward in time. The Jacobian matrix $\boldsymbol{M}$ comes with a natural co-ordinate basis, $\{\partial/\partial x^i\}$. By constructing another, more convenient basis, through the density matrix the dynamics are governed by $\boldsymbol{P}$. That $\boldsymbol{P}$ is similar to $\boldsymbol{M}$, implies $\boldsymbol{P} \to \boldsymbol{M}$ when the density matrix is expressed in the coordinate basis. The contravariant metric tensor $\boldsymbol{g}_{\boldsymbol{\xi}}^{-1}$ transform as:

$$\boldsymbol{g}_{\boldsymbol{\xi}}^{-1}(t) = \boldsymbol{\Psi'}^{\top}\boldsymbol{\Psi'} = \boldsymbol{P}^{\top}\boldsymbol{\Psi}^{\top}\boldsymbol{\Psi}\boldsymbol{P} = \boldsymbol{P}^{\top}\boldsymbol{g}_{\boldsymbol{\xi}}^{-1}(t_0)\boldsymbol{P}, \tag{B8}$$

and the covariant metric tensor $\boldsymbol{g}_{\boldsymbol{\xi}}(t)$ transforms as:

$$\boldsymbol{g}_{\boldsymbol{\xi}}(t) = \boldsymbol{P}^{-1}\boldsymbol{g}_{\boldsymbol{\xi}}(t_0)\boldsymbol{P}^{-\top}. \tag{B9}$$

It follows from these relationships that $\boldsymbol{\xi}$ and $\boldsymbol{g}_{\boldsymbol{\xi}}^{-1}$ are similar:

$$\boldsymbol{\Psi'}^{-1}\,\boldsymbol{\xi}(t)\,\boldsymbol{\Psi'} = \boldsymbol{\Psi'}^{-1}\,\boldsymbol{\Psi'}\boldsymbol{\Psi'}^{\top}\,\boldsymbol{\Psi'} = \boldsymbol{g}_{\boldsymbol{\xi}}^{-1}(t). \tag{B10}$$

If $\boldsymbol{\Psi'} = \boldsymbol{M}\boldsymbol{\Psi} = \boldsymbol{\Psi}\boldsymbol{P}$ with $\boldsymbol{P} = \boldsymbol{\Psi}^{-1}\boldsymbol{M}\boldsymbol{\Psi}$ then

$$\boldsymbol{g}_{\boldsymbol{\xi}}(t)^{-1} = \boldsymbol{\Psi'}^{-1}\,\boldsymbol{\xi}(t)\,\boldsymbol{\Psi'} = \boldsymbol{P}^{-1}\boldsymbol{g}_{\boldsymbol{\xi}}^{-1}(t_0)\boldsymbol{P}. \tag{B11}$$

The linear independence of the basis vectors in $\boldsymbol{\Psi}$ guarantees it is invertible.

For a general Riemannian manifold, the volume n-form determines the *invariant* volume element $d\tilde{\mathcal{V}}$ in an arbitrary coordinate system: $d\tilde{\mathcal{V}} = \sqrt{g_{\boldsymbol{\xi}}}d\mathcal{V}$, where $g_{\boldsymbol{\xi}}$ is the determinant of the covariant metric tensor $\boldsymbol{g}_{\boldsymbol{\xi}}$. From Eq. B10, $\boldsymbol{g}_{\boldsymbol{\xi}}$ is similar to $\boldsymbol{\xi}^{-1}$.

Furthermore, Eq. 5 provides the compatibility condition of the metric tensor with the flow:

$$\begin{aligned}\frac{d}{dt}\ln|\boldsymbol{\xi}| &= 2\,\boldsymbol{\nabla}\cdot\dot{\boldsymbol{x}}\\ \frac{d}{dt}|\boldsymbol{\xi}| &= 2|\boldsymbol{\xi}|\boldsymbol{\nabla}\cdot\dot{\boldsymbol{x}}\\ \frac{1}{2}|\boldsymbol{\xi}|^{-\frac{3}{2}}\frac{d}{dt}|\boldsymbol{\xi}| &= |\boldsymbol{\xi}|^{-\frac{1}{2}}\boldsymbol{\nabla}\cdot\dot{\boldsymbol{x}}\\ \frac{d}{dt}|\boldsymbol{\xi}|^{-\frac{1}{2}} &= -|\boldsymbol{\xi}|^{-\frac{1}{2}}\boldsymbol{\nabla}\cdot\dot{\boldsymbol{x}}.\end{aligned} \tag{B12}$$

We can express this relation in terms of the metric determinant [38]:

$$\begin{aligned}\frac{d}{dt}\sqrt{g_{\boldsymbol{\xi}}} &= -\sqrt{g_{\boldsymbol{\xi}}}\boldsymbol{\nabla}\cdot\dot{\boldsymbol{x}}\\ \frac{\partial\sqrt{g_{\boldsymbol{\xi}}}}{\partial t} + \dot{\boldsymbol{x}}\cdot\boldsymbol{\nabla}\sqrt{g_{\boldsymbol{\xi}}} &= -\sqrt{g_{\boldsymbol{\xi}}}\boldsymbol{\nabla}\cdot\dot{\boldsymbol{x}}\\ \frac{\partial}{\partial t}\sqrt{g_{\boldsymbol{\xi}}} + \boldsymbol{\nabla}\cdot(\sqrt{g_{\boldsymbol{\xi}}}\dot{\boldsymbol{x}}) &= 0.\end{aligned}$$

Replacing $g_{\boldsymbol{\xi}}$ by $|\boldsymbol{\xi}|^{-1}$, we obtain the generalized Liouville equation:

$$\frac{\partial}{\partial t}(|\boldsymbol{\xi}|^{-\frac{1}{2}}) + \boldsymbol{\nabla}\cdot(|\boldsymbol{\xi}|^{-\frac{1}{2}}\dot{\boldsymbol{x}}) = 0. \tag{B13}$$

For Hamiltonian dynamics, the metric is time-independent because of the vanishing flow divergence,

$$\frac{\partial}{\partial t}(|\boldsymbol{\xi}|^{-\frac{1}{2}}) + \dot{\boldsymbol{x}}\cdot\boldsymbol{\nabla}|\boldsymbol{\xi}|^{-\frac{1}{2}} = 0. \tag{B14}$$

By introducing a norm-preserving dynamics and a normalized density matrix, the form of the generalized Liouville equation is identical to the traditional Liouville equation. Defining the metric determinant $g_{\boldsymbol{\varrho}} = |\boldsymbol{\varrho}|^{-1}$, the conservation of the normalized density matrix $\boldsymbol{\varrho}$,

$$\frac{d}{dt}\ln|\boldsymbol{\varrho}| = 0,$$

is equivalent to the generalized Liouville equation:

$$\begin{aligned} \frac{d}{dt}(|\boldsymbol{\varrho}|^{-\frac{1}{2}}) &= \frac{d}{dt}\sqrt{g_{\boldsymbol{\varrho}}} = 0 \\ \frac{\partial}{\partial t}(|\boldsymbol{\varrho}|^{-\frac{1}{2}}) + \dot{\boldsymbol{x}}\cdot\boldsymbol{\nabla}|\boldsymbol{\varrho}|^{-\frac{1}{2}} &= 0 \qquad \text{(B15)} \\ \frac{\partial}{\partial t}(\sqrt{g_{\boldsymbol{\varrho}}}) + \dot{\boldsymbol{x}}\cdot\boldsymbol{\nabla}\sqrt{g_{\boldsymbol{\varrho}}} &= 0. \qquad \text{(B16)} \end{aligned}$$

The Liouville equations for $\boldsymbol{\xi}$ and $\boldsymbol{\varrho}$ are related. Taking the determinant of Eq. 15, we find

$$\begin{aligned} |\boldsymbol{\varrho}| &= \frac{|\boldsymbol{\xi}|}{(\mathrm{Tr}\,\boldsymbol{\xi})^n} \\ g_{\boldsymbol{\varrho}} &= g_{\boldsymbol{\xi}}\,(\mathrm{Tr}\,\boldsymbol{\xi})^n \\ \sqrt{g_{\boldsymbol{\varrho}}} &= \sqrt{g_{\boldsymbol{\xi}}}\,(\mathrm{Tr}\,\boldsymbol{\xi})^{\frac{n}{2}} \\ \ln\sqrt{g_{\boldsymbol{\varrho}}} &= \ln\sqrt{g_{\boldsymbol{\xi}}} + \frac{n}{2}\ln(\mathrm{Tr}\,\boldsymbol{\xi}). \qquad \text{(B17)} \end{aligned}$$

Recalling that $g_{\boldsymbol{\varrho}}$ is time-independent, the ratio $|\boldsymbol{\xi}|/\,\mathrm{Tr}\,\boldsymbol{\xi}^n$ is a constant of motion for any dynamical system.

It is also possible to express the compatibility condition using the trace of $\boldsymbol{g}_{\boldsymbol{\xi}}^{-1}$,

$$-\frac{d}{dt}\ln\sqrt{g_{\boldsymbol{\xi}}} = \frac{n}{2}\frac{d}{dt}\ln(\mathrm{Tr}\,\bar{\boldsymbol{g}}_{\boldsymbol{\xi}}) = \boldsymbol{\nabla}\cdot\dot{\boldsymbol{x}}. \tag{B18}$$

For the normalized density matrix $\boldsymbol{\varrho}$ of the form $\boldsymbol{\varrho}(t) = n^{-1}\sum_{i=1}^{n}\boldsymbol{\varrho}_i(t)$, we have averages similar to those in quantum mechanics. For example, the average

$$\begin{aligned} \mathrm{Tr}(\boldsymbol{A}_+\boldsymbol{\varrho}) &= n^{-1}\sum_{i=1}^{n}\mathrm{Tr}(\boldsymbol{A}_+\boldsymbol{\varrho}_i) \\ &= n^{-1}\sum_{i=1}^{n} r_i = n^{-1}\,\mathrm{Tr}\,\boldsymbol{A}_+, \qquad \text{(B19)} \end{aligned}$$

where r_i is the instantaneous Lyapunov exponent for the i^{th} basis state. This average is related to the divergence of the flow: $\boldsymbol{\nabla}\cdot\dot{\boldsymbol{x}} = n\,\mathrm{Tr}(\boldsymbol{A}_+\boldsymbol{\varrho})$.

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