Dissipative signatures of dynamical phases and transitions

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Abstract

Living and non-living active matter consumes energy at the microscopic scale to drive emergent, macroscopic behavior including traveling waves and coherent oscillations. Recent work has characterized non-equilibrium systems by their total energy dissipation, but little has been said about how dissipation manifests in distinct spatiotemporal patterns. We introduce a novel measure of dissipation we term the entropy production factor (EPF) to quantify how time reversal symmetry is broken in field theories across scales. We illustrate the use of this method on simulations of the Brusselator, a prototypical biochemically motivated non-linear oscillator. We find that while the total dissipation does not drastically change as the system undergoes a Hopf bifurcation, the EPF shows a dramatic change in the allocation of energy. The EPF measures how microscopic irreversibility propagates to larger length and time scales and integration through frequency space bounds the net dissipation of stochastic processes.

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I. INTRODUCTION

In many-body systems, collective behavior that breaks time-reversal symmetry can emerge due to the consumption of energy by the individual constituents [1–3]. In biological, engineered, and other naturally out of equilibrium processes, entropy must be produced so as to bias the system in a “forward” direction [4–9]. How this microscopic breaking of time reversal symmetry manifests at larger length and time scales in different ways remains unclear. For example, bulk order parameters in complex reactions can switch from exhibiting incoherent, disordered behavior to stable static patterns [10, 11] or traveling waves of excitation [12, 13] that break time reversal symmetry in both time and space simply by altering the strength of the microscopic driving. Recent advances in stochastic thermodynamics have highlighted the use of entropy production as a quantity to measure a system’s distance from equilibrium [14–18]. However, there has been conflicting evidence concerning the continuity of entropy production rates and its derivatives at these transitions, especially when small molecule numbers or spatial dynamics are involved [19–21]. To date, it is unknown the extent to which complex spatiotemporal patterns can be uniquely specified by a net dissipation rate.

To address this, we introduce what we term the entropy production factor (EPF), a dimensionless function of frequency and wave-vector. The EPF must be zero in equilibrium and is greater than zero out of equilibrium, quantifying the time and length scales at which non-equilibrium behavior manifests. Integrating the EPF produces a loose bound on the entropy production rate (EPR) of a system. We illustrate how to calculate the EPF directly from data using the analytically tractable example of coupled Gaussian fields obeying Model A dynamics [22]. We then turn to the Brusselator reaction-diffusion model for spatiotemporal biochemical oscillations to study the connections between pattern formation and dissipation. Perhaps surprisingly, dynamical transitions which separate qualitatively different macroscopic behavior are not always accompanied by a change in the EPR. However, they do leave dramatic traces in the EPF. Importantly, the EPF can be calculated in any number of spatial dimensions, making it broadly applicable to a wide variety of data types, from particle tracking to 3+1 dimensional microscopy time series.
II. ENTROPY PRODUCTION FACTOR DERIVATION

Consider a system described by a set of \( M \) real, random variables obeying some possibly unknown dynamics. A specific trajectory of the system over a total time \( T \) is given by \( X = \{ X^i(t) | t \in [0, T] \} \). Given an ensemble of trajectories, the average EPR, \( \dot{S} \equiv \langle dS/dt \rangle \), is given by \[ 5, 6, 23 \]

\[
\dot{S} = \lim_{T \to \infty} \frac{1}{T} D_{KL} \left( P[X], \tilde{P}[X] \right); \quad D_{KL} \left( P[X], \tilde{P}[X] \right) = \left\langle \log \left( \frac{P[X]}{\tilde{P}[X]} \right) \right\rangle_p
\]

where we have set \( k_B = 1 \) throughout and \( D_{KL} \) denotes the Kullback-Leibler divergence between two probability distributions. \( P[X] \) and \( \tilde{P}[X] \) are the probability distribution functionals of observing the path \( X(t) \) of length \( T \) and the probability of observing its reverse path, respectively. Writing the Fourier transform of \( X^i(t) \) as \( x^i(\omega) \) and writing the column vector \( x(\omega) = (x^1(\omega), x^2(\omega), \ldots)^T \), we assume the paths obey a Gaussian distribution:

\[
P[x(\omega)] = \frac{1}{Z} \prod_{\omega_n} \exp \left( -\frac{1}{2T} x^\dagger C^{-1} x \right),
\]

where \( x^\dagger \) denotes the conjugate transpose of the vector \( x \) evaluated at the discrete frequencies \( \omega_n = 2\pi n/T \). \( C(\omega_n) \) is the covariance matrix in Fourier space with elements \( C^{ij}(\omega_n) = \langle x^i(\omega_n)x^j(-\omega_n) \rangle / T \), and \( Z \) is the partition function. The expression for \( \tilde{P}[x] \) is identical but with \( C^{-1}(\omega_n) \rightarrow C^{-1}(-\omega_n) \) (see Supplementary Material). Combining Eq. 1 with Eq. 2 and taking \( T \to \infty \), we arrive at our main result:

\[
\dot{S} = \int \frac{d\omega}{2\pi} \mathcal{E}(\omega); \quad \mathcal{E}(\omega) = \frac{1}{2} \left[ C^{-1}(-\omega) - C^{-1}(\omega) \right]_{ij} C^{ji}(\omega).
\]

This defines the EPF, \( \mathcal{E}(\omega) \), which measures rotations between \( M \geq 2 \) variables in phase space. Integrating \( \mathcal{E} \) gives the EPR, \( \dot{S} \), in this Gaussian approximation. For a more complex system Eq. 3 gives an approximate bound; some entropy production may be invisible in the dynamics of \( X \) and some asymmetry in \( X \) may only appear in its higher moments. As \( C^{ii}(\omega) = C^{ii}(-\omega) \) the only contributions to \( \mathcal{E} \) come from the cross-covariances between the random variables of interest. As such, this bound yields exactly 0 for a single variable.

This formulation extends naturally to random fields. For \( M \) random fields \( \phi = \{ \phi^i(x, t) | t \in [0, T], x \in \mathbb{R}^d \} \) and in \( d \) spatial dimensions the EPR density, \( \dot{s} \equiv \dot{S}/V \) where \( V \) is the system volume, becomes [See Supplementary Material]:

\[
\dot{s} = \int \frac{d\omega}{2\pi} \frac{d^d q}{(2\pi)^d} \mathcal{E}(q, \omega); \quad \mathcal{E}(q, \omega) = \frac{1}{2} \left[ C^{-1}(q, -\omega) - C^{-1}(q, \omega) \right]_{ij} C^{ji}(q, \omega).
\]
where \( C^{ij}(q, \omega) \) is the structure factor and where \( \mathcal{E}(q, \omega) \), is now a function of wavevector and frequency. Here, we neglect the term \( \ln(\tilde{Z}/Z)/T \) which can be non-zero only in systems that lack both parity and rotational symmetry [see Supplementary Material].

We calculate \( \mathcal{E} \) from data by estimating covariance functions, or cross-spectral densities [24]. To use Eq. 4, we consider data of \( N \) finite length trajectories of \( M \) variables over a time \( T \) in \( d \) spatial dimensions. Each dimension has a length \( L_i \). We create an estimate of the covariance matrix, \( \tilde{C}(q, \omega) \), from time-series using standard methods [see Methods]. These measurements will inevitably contain noise that is not necessarily time-reversal symmetric, even for an equilibrium system. Noise in the estimate of \( \tilde{C} \) from a single experiment (\( N = 1 \)) will systematically bias our estimated \( \mathcal{E} \) by \( \Delta \mathcal{E} = M(M-1)/2 \) at each frequency and will thereby introduce bias and noise to the measurement of \( \dot{s} \). We can simply remove the bias from our measured \( \mathcal{E} \), but to reduce the variance, we smooth \( \tilde{C} \) by component-wise convolution with a multivariate Gaussian of width \( \sigma = (\sigma_{q_1}, \ldots, \sigma_{q_d}, \sigma_\omega) \) in frequency space, giving \( \hat{C} \). This is equivalent to multiplying each component of the time domain \( \tilde{C}(r, t) \) by a Gaussian, cutting off long noisy tails in the real space covariance functions. We can then use \( \hat{C} \) in Eq. 4 to create our final estimator for the EPF, \( \hat{\mathcal{E}} \), and thereby the EPR, \( \hat{\dot{s}} \). We calculate and remove the bias in \( \hat{\mathcal{E}} \) and \( \hat{\dot{s}} \) in all results below [see Methods].

III. RESULTS

To illustrate the information contained in \( \mathcal{E} \), its numerical estimation, and the accuracy of \( \dot{s} \), we analyze simulations of coupled, 1 dimensional Gaussian stochastic fields for which \( \mathcal{E} \) and \( \dot{s} \) can be calculated analytically. We then study simulations of the reaction-diffusion Brusselator, a prototypical model for non-linear biochemical oscillators, and use \( \mathcal{E} \) to study how dissipation manifests as broken detailed balance at different time and length scales as the system undergoes a Hopf bifurcation [25].
A. Driven Gaussian fields

Consider two fields obeying Model A dynamics [22] with non-equilibrium driving parametrized by $\alpha$:

$$\begin{align*}
\partial_t \phi(x,t) &= -D \frac{\delta F}{\delta \phi} - \alpha \psi + \sqrt{2D} \xi \\
\partial_t \psi(x,t) &= -D \frac{\delta F}{\delta \psi} + \alpha \phi + \sqrt{2D} \xi,
\end{align*}$$

(5)

where $\xi(x,t)$ is Gaussian white noise with variance $\langle \xi_i(x,t) \xi_j(x',t') \rangle = \delta_{ij} \delta(x-x') \delta(t-t')$, $D$ is a relaxation constant, and $\delta F/\delta \phi$ is the functional derivative with respect to $\phi$ of the free energy $F$ given by:

$$F = \int dx \left[ \frac{r}{2} (\phi^2 + \psi^2) + \frac{1}{2} (|\partial_x \phi|^2 + |\partial_x \psi|^2) \right],$$

(6)

so that the fields have units of $\ell^{1/2}$ and $r$ penalizes large amplitudes.

The EPR density, $\dot{s}$, is calculated analytically in two ways. First, we solve Eq. 1 directly using the Onsager-Machlup functional for the path probability functional of $\eta(x,t) = (\phi(x,t), \psi(x,t))^T$ [4, 26]. Second, the covariance matrices are calculated analytically, used to find $E$ through Eq. 4, and integrated to find $\dot{s}$. Both cases give the same result for $\dot{s}$. The result for both $E$ and $\dot{s}$ are [Supplementary Material]:

$$\begin{align*}
E^{DGF} &= \frac{8\alpha^2 \omega^2}{(\omega^2 - \omega_0^2(q))^2 + (2D(r + q^2)\omega)^2}, \\
\dot{s}^{DGF} &= \frac{\alpha^2}{D \sqrt{r}}.
\end{align*}$$

(7)

We see that $E^{DGF} \geq 0$ and exhibits a peak at $(q, \omega) = (0, \omega_0(0))$, where $\omega_0(q) = \sqrt{(D(r + q^2))^2 + \alpha^2}$, indicating that the system is driven at all length scales with a driving frequency of $\alpha$, dampened by an effective spring constant $Dr$. In addition, it is clear that multiple combinations of $\alpha$, $r$, and $D$ can give the same value for $\dot{s}$ while $E$ distinguishes between equally dissipative trajectories in the shape and location of its peaks. In this way, $E$ gives information about the form of the underlying dynamics not present in the total EPR.

We perform simulations to assess how well $E$ can be extracted from time series data of fields [See methods for details]. The estimated $\hat{E}$ shows excellent agreement with Eq. 7 (Fig. 1). Integrating $\hat{E}$ gives $\hat{s}$, which also shows good agreement with $\dot{s}^{DGF}$.

Our estimator gives exact results for the driven Gaussian fields because the true path probability functional for these fields is indeed Gaussian. In contrast, the complex patterns seen in nature arise from systems obeying highly non-linear dynamics. For such dynamics,
FIG. 1. Entropy production rate and entropy production factor are well estimated for driven Gaussian fields. (a) Snapshot of typical configurations of both fields, \((\psi, \phi)\) obeying Eq. 5 for \(\alpha = 7.5\). (b) Subsection of a typical trajectory for one field for \(\alpha = 7.5\) in dimensionless units. Colors indicate the value of the field at each point in spacetime. (c) \(\hat{\mathcal{E}}\) for \(\alpha = 7.5\). Contours show level sets of \(\mathcal{E}^{DGF}\). (d) Measured \(\hat{s}\) vs. \(\alpha\) for simulations of total time \(T = 50\) and length \(L = 12.8\). Red line shows the theoretical value, \(s^{DGF}\). Mean \(\pm\) standard deviation of \(\hat{s}\) given by black dots and shaded area. See Supplementary Material for all simulation parameters.

our Gaussian approximation is no longer exact. To investigate how energy dissipation correlates with pattern formation, we study simulations of the Brusselator model for biochemical oscillations [27]. We begin by describing the various dynamical phases of the equations of motion. Next, we calculate \(\mathcal{E}\) and \(\hat{S}\) for only the reactions before adding diffusion to study the synchronized oscillations that arise in the 1 dimensional reaction-diffusion system.

**B. Reaction-diffusion Brusselator**

We use a reversible Brusselator model [27, 28] with dynamics governed by the reaction equations:

\[
A \xrightleftharpoons[k_1^-]{k_1^+} X; \quad B + X \xrightleftharpoons[k_2^-]{k_2^+} Y + C; \quad 2X + Y \xrightleftharpoons[k_3^-]{k_3^+} 3X; \tag{8}
\]

where \(A\), \(B\), and \(C\) are fixed. The system is in equilibrium when the external chemical baths and reaction rates obey \(Bk_2^+k_3^+ = Ck_2^-k_3^-\). When this equality is violated, the system is driven away from equilibrium and exhibits cycles in the \((X,Y)\) plane. Defining

\[
\Delta \mu = \ln \left( \frac{Bk_2^+k_3^+}{Ck_2^-k_3^-} \right), \tag{9}
\]
FIG. 2. Brusselator dynamics exhibit circulation without macroscopic oscillatory solution. (a) Eigenvalues of the Brusselator’s relaxation matrix, $R$ as a function of $\Delta \mu$. $\lambda_{\pm}$ shown in red and blue, respectively, with each color going from dark to light with increasing $\Delta \mu$. The red and blue arrows serve as guides for the reader to follow the trajectory of $\lambda_{\pm}$. With our parameters, the stable focus appears at $\Delta \mu = 5.26$ and the Hopf bifurcation occurs at $\Delta \mu_{HB} = 6.16$. (b) Probability distributions (blue) and probability fluxes (red arrows) for Brusselator simulations with $\Delta \mu = [-1, 0, 1]$, showing the reversal in flux circulation direction at $\Delta \mu = 0$.

The Brusselator is at equilibrium when $\Delta \mu = 0$ and is driven into a non-equilibrium steady state when $\Delta \mu \neq 0$. We vary $B$ and $C$ to change $\Delta \mu$ while keeping the product $(Bk_2^+k_3^+)(Ck_2^-k_3^-)$ constant, keeping the rate at which reactions occur constant for all $\Delta \mu$ [29].

As $\Delta \mu$ increases, the Brusselator undergoes dynamical phase transitions. For all $\Delta \mu$, there exists a steady state $(X_{ss}, Y_{ss})$, the stability of which is determined by the relaxation matrix, $R$, of the macroscopic equations corresponding to Eq. 8 (Fig. 2) [Supplementary Material]. The two eigenvalues of $R$, $\lambda_{\pm}$, divide the steady state into four classes [25]:

1. $\lambda_{\pm} \in \mathbb{R}_{<0} \rightarrow$ Stable attractor, no oscillations

2. $\lambda_{\pm} \in \mathbb{C}$, $\text{Re}[\lambda_{\pm}] < 0 \rightarrow$ Stable focus
3. \( \lambda_\pm \in \mathbb{C}, \quad \text{Re}[\lambda_\pm] > 0 \rightarrow \text{Hopf Bifurcation, limit cycle} \)

4. \( \lambda_\pm \in \mathbb{R}_{>0} \rightarrow \text{Unstable repeller} \)

The eigenvalues undergo these changes as \( \Delta \mu \) changes, allowing us to consider \( \Delta \mu \) as a bifurcation parameter. We define \( \Delta \mu_{HB} \) as the value of \( \Delta \mu \) where the system undergoes the Hopf bifurcation.

Non-equilibrium steady states are traditionally characterized by their circulation in a phase space \([30–33]\). One may then question how it is possible to detect non-equilibrium effects in the Brusselator around \( \Delta \mu = 0 \) when the system’s steady state is a stable attractor with no oscillatory component. While this is true for the macroscopic dynamics used to derive \( \lambda_\pm \), we simulate a system with finite numbers of molecules subject to fluctuations. These stochastic fluctuations give rise to circulating dynamics, even when the deterministic dynamics do not \([28]\). Indeed, we see persistent circulation in the \((X,Y)\) plane when \( \lambda_\pm \in \mathbb{R}_{<0} \), with the vorticity changing sign around \( \Delta \mu = 0 \) (Fig. 2b).

In order to assess the accuracy of our estimated EPR, \( \hat{S} \), we calculate an estimate of the true EPR, \( \dot{S}_{\text{true}} \), for a simulation of Eq. 8 by calculating the exact entropy produced by each reaction that occurs in the trajectory \([34]\), and then fitting a line to the cumulative sum to estimate \( \dot{S}_{\text{true}} \) (Supplementary Figure 1, Methods). We find that \( \hat{S} \) significantly underestimates \( \dot{S}_{\text{true}} \) (note the logged axes in Fig. 3). This has a simple yet significant cause. Our method relies purely on system dynamics to give \( \hat{S} \). However, multiple reactions in Eq. 8 lead to the same dynamics, despite those reactions causing different amounts of dissipation. In order to account for this, we recalculate \( \hat{S} \) by summing over all reactions that could give rise to the change in \((X,Y)\) seen at each time point [see Methods]. This “blinded” estimate of the EPR, \( \dot{S}_{\text{blind}} \), shows excellent agreement with \( \hat{S} \), despite the highly non-linear dynamics of Eq. 8. All three estimates of \( \dot{S} \) are quadratic around \( \Delta \mu = 0 \) (Supplementary Figure 2).

One may expect \( \dot{S} \), which itself depends on \( \Delta \mu \), to reflect changes in the stability of the steady state. Indeed, suggesting the emergence of an oscillatory steady state would be ripe for analysis using \( \dot{S} \). However, \( \dot{S} \) varies smoothly through this transition (Fig. 3). Further from equilibrium, \( \dot{S} \) begins to underestimate \( \dot{S}_{\text{blind}} \) when the steady state becomes a stable focus, following a shape similar to previous studies of entropy production in stochastic simulations of biochemical oscillators \([19]\). This underestimate is also present in a measure of
FIG. 3. Brusselator entropy production rate changes smoothly through Hopf bifurcation. (a) Typical trajectory in \((X, Y)\) space for \(\Delta \mu = 6.2\). The occupation probability distribution is shown in blue, with a subsection of a typical trajectory shown in black. The end of the trajectory is marked by the white circle. Inset shows the same information for the system at equilibrium, where \(\Delta \mu = 0\), with the same colorbar as the main figure. (b) \(\dot{\mathcal{E}}\) for \(\Delta \mu = [3.5, 5.3, 6.2]\) shown in green, orange, and purple, respectively. Shaded area shows mean ± std of \(\dot{\mathcal{E}}\) for \(N = 50\) simulations. Inset shows the same curves on a log-log scale. (c) \(\dot{S}\) as a function of \(\Delta \mu\). Blue, orange, and black points show results for \(\dot{S}_{\text{true}}, \dot{S}_{\text{blind}},\) and \(\dot{\mathcal{S}}\), respectively. Shaded area shows mean ± std of \(\dot{\mathcal{S}}\) for \(N = 50\) simulations. See Supplementary Material for all simulation parameters.

\(\dot{\mathcal{S}}\) using phase space fluxes in a Fokker-Planck approximation of the dynamics [6, 16, 17, 31, 32] (Supplementary Figure 2), reflecting the linear approximation made in deriving Eq. 3.

For \(\Delta \mu < \Delta \mu_{HB}\), \(\dot{\mathcal{E}}\) exhibits a single peak that increases in amplitude while decreasing in frequency as \(\Delta \mu\) increases. Above \(\Delta \mu_{HB}\), additional peaks at integer multiples of the peak frequency appear due to the non-linear shape of the limit cycle attractor. These harmonics are expected for dynamics on a non-circular path. We conclude that the Hopf bifurcation does not have any obvious signature in the dissipation of the Brusselator reactions alone.

To investigate how dynamical phase transitions manifest in the dissipation of spatially extended systems, we simulate a reaction-diffusion Brusselator on a 1 dimensional periodic lattice with \(L\) compartments spaced a distance \(h\) apart. The full set of reactions are now

\[
\begin{align*}
A_i &\xrightleftharpoons[k_1^{-1}][k_1^+] X_i; & B_i + X_i &\xrightarrow[k_2^{-1}][k_2^+] Y_i + C_i; & 2X_i + Y_i &\xrightarrow[k_3^{-1}][k_3^+] 3X_i; \\
X_i &\xrightleftharpoons[d_X] X_{i+1}; & Y_i &\xrightleftharpoons[d_Y] Y_{i+1}; & i \in [1, L]
\end{align*}
\]
FIG. 4. 1 dimensional reaction-diffusion Brusselator network shows emergent collective behavior above Hopf Bifurcation with smooth increase in $\dot{S}$. (a) Subsection of a typical trajectory for $X(r,t)$ and $Y(r,t)$ for (a) $\Delta \mu = 3.5$, below the Hopf Bifurcation and (b) $\Delta \mu = 6.2$, above it. Color indicates the local number of the chemical species. (b) $\dot{s}$ as a function of $\Delta \mu$. Blue, orange, and black points show results, respectively. Shaded area shows $\pm 1 \sigma$ of $N = 10$ simulations. Red dashed line shows location of $\Delta \mu_{HB}$. See Supplementary Material for all simulation parameters.

where $d_j = D_j/h^2$, and $D_j$ is the diffusion constant of chemical species $j = [X,Y]$. Qualitatively different dynamics occur based on the ratio $D_X/D_Y$. $D_X/D_Y \ll 1$ yields static Turing patterns. We focus on the $D_X/D_Y \gg 1$ regime which exhibits dynamic, excitable waves. All values of $\{A_i, B_i, C_i\}$ are kept constant in each compartment.

In the absence of noise, the reaction-diffusion Brusselator has the same dynamics as the well mixed Brusselator, and so it is not surprising that it’s EPR curve as a function of $\Delta \mu$ is similar. However, unlike the well-mixed system, the Hopf bifurcation signals the onset of qualitatively distinct dynamics in the reaction-diffusion system. Prior to the Hopf bifurcation, there are no coherent, spatial patterns in the system’s dynamics (Fig. 4a). Above the Hopf bifurcation, the system dynamics change drastically. Rather than each lattice site behaving roughly independently, system-spanning waves begin to emerge that synchronize the oscillations across the system (Fig. 4b).

Throughout these changes, the system is driven further from equilibrium, as reflected in the increasing $\dot{s}$ (Fig. 4c). The shift to collective behavior is not accompanied by a dramatic shift in $\dot{S}$, as was the case with only the Brusselator reactions. Instead, $E$ carries
FIG. 5. Entropy production factor highlights macroscopic behavior after undergoing a Hopf bifurcation in the reaction-diffusion Brusselator model. (a) $\langle E \rangle$ over $N = 10$ simulations for $\Delta \mu = 3.5$, i.e. $\Delta \mu < \Delta \mu_{HB}$. (b) $\langle E \rangle$ over $N = 10$ simulations for $\Delta \mu = 6.2$, i.e. $\Delta \mu > \Delta \mu_{HB}$. Same conditions as Fig. 4. (c) Wavenumber, $q$, that maximizes $\hat{E}$ as a function of $\Delta \mu$. Red line shows $\Delta \mu_{HB}$. Inset shows zoom in of $\hat{s}$ around $\Delta \mu_{HB}$.

the signature of the dynamical phase transition. For $\Delta \mu < \Delta \mu_{HB}$, $\hat{E}$ shows peaks at high wavenumbers, reflecting that dissipation is occurring incoherently over short length scales. Above $\Delta \mu_{HB}$, as the system shows synchronized oscillations, there is an abrupt shift in the peaks of $\hat{E}$ to low $q$, indicating that this collective behavior carries the majority of the dissipation (Fig. 5b,c). We also infer that the collective behavior is partially composed of traveling waves due to the streaks in $\hat{E}$ (Fig. 5b).

IV. DISCUSSION

We have introduced a novel quantity, the entropy production factor $\mathcal{E}$, that quantifies dissipation in macroscopic, time-reversal symmetry breaking behavior driven by the microscopic consumption of energy. $\mathcal{E}$ measures energy dissipated by the interactions between multiple variables through the measurement of time-reversal symmetry breaking in their cross-covariances. There is no need for external probes to measure a response function, in contrast to methods based on the Harada-Sasa equality [26, 35], because $\mathcal{E}$ is concerned with the physics of non-equilibrium, many-body interactions rather than how a single variable breaks the fluctuation-dissipation theorem. Integrating $\mathcal{E}$ gives a loose bound on the net entropy production rate, $\dot{s}$. Better estimates can be achieved by including higher cumulants.
in the action of Eq. 2. We believe that these higher-order terms will contribute positively to $\dot{s}$, making our estimate a lower-bound.

Here, we illustrated that a total dissipation rate does not reflect the transition to coherent oscillations in simulations of the reaction-diffusion Brusselator system as it undergoes a Hopf bifurcation, similarly to what was seen recently for transitions to Turing patterns [21], but is instead present in $\mathcal{E}$ as energy dissipation manifests at long length-scales. This is consistent with results from reconstituted actomyosin networks [36] and cyanobacterial circadian clocks [37, 38] that transition to qualitatively distinct behaviors (long-range contractility and oscillator synchronization, respectively) while the total dissipation changes smoothly through the transition. All together, these results suggest that the distribution of dissipation, not its sum, can uniquely characterize dynamical phase transition in non-equilibrium field theories.

Calculating $\mathcal{E}$ does not require knowledge about the form of the underlying dynamics and is easy to calculate for many types of data, including both random variables, such as the positions of driven colloidal particles [39], and random fields, such as spatially heterogeneous protein concentrations in cells [40]. While the examples considered here are simulations of 1+1 dimensional fields, there is nothing inherently different in the methodology used here if one were to analyze experimental data in 2 or 3 spatial dimensions, such as the 3+1 dimensional time series data attained using lattice-light sheet microscopy [41]. As techniques for imaging active living and non-living matter continue to give more sophisticated, spatially resolved data, measuring dissipation in multi-scale processes via $\mathcal{E}$ will provide more nuanced information about the underlying non-equilibrium processes than is contained the total entropy production rate.

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VI. METHODS

A. Calculating $\mathcal{E}$ from data

Estimate $\mathcal{E}$ requires estimating frequency-space covariance functions, or cross spectral densities (CSDs). Considering a set of $M$ discrete, real variables measured over time: \( \{X^i(t)\} \), where $t = \Delta t, \ldots, T$, with $T = N \Delta t$, and $i = 1, \ldots, M$ indexes the variables, we estimate the CSD using the periodogram,

$$
\tilde{C}_{ij}(\omega_n) = \frac{1}{N^2} x^i(\omega_n) x^j(-\omega_n) \tag{11}
$$

where $x^i(\omega) = \mathcal{F}\{X^i(t) - (X^i(t))\}$ are the Fourier transforms of the centered variables over the frequencies $\omega_n = \frac{2\pi n}{T}$ for $n = [-N/2, N/2]$.

The periodogram, is known to exhibit a systematic bias and considerable variance in estimating the true CSD. Both of these issues can be resolved by smoothing $\tilde{C}_{ij}$ via convolution with a Gaussian with width $\sigma$. This is equivalent to multiplying $\tilde{C}_{ij}$ in the time domain by a Gaussian of width $1/\sigma$. We then define our smoothed CSD as

$$
\hat{C}_{ij}(\omega_n) = \sum_{\omega_\mu} \Delta \omega \frac{\exp[-(\omega_\mu - \omega_n)^2/2\sigma^2]}{\sqrt{2\pi \sigma^2}} \tilde{C}_{ij}(\omega_\mu) \tag{12}
$$

Once $\hat{C}$ is calculated, we then use the discrete version of Eq. 3 to estimate $\mathcal{E}$. The extension to higher-dimensional data is done as follows: taking into account the spatial lattice on which the data is taken in Eq. 11, convolving the result with a multivariate Gaussian in Eq. 12, and finally estimate $\dot{s}$ using the discrete version of Eq. 4.

B. Bias in $\hat{\mathcal{E}}$ and $\hat{S}$

Our estimates of $\hat{\mathcal{E}}$ and $\hat{S}$ are biased. The bias is found by calculating the expected value of $\hat{S}$ for a system in equilibrium. To do this, we assume that the true covariance function is $C^{ij} = \delta^{ij}$ and measurement noise plus finite sampling time and rate gives rise to Gaussian noise in both the real and complex parts of $\tilde{C}^{ij}(\omega)$, obeying the symmetries required for $C^{ij}$
to be Hermitian. We only cite the results here and refer the reader to the Supplementary Material for a full derivation. The bias for random variables is

$$E_{\text{bias}} = \frac{M(M - 1)}{2} \sqrt{\pi \over T\sigma}$$

$$\dot{E}_{\text{bias}} = \frac{M(M - 1)}{2} \frac{\omega_{\text{max}}}{T\sigma \sqrt{\pi}}$$

(13)

where $M$ is the number of variables, $\omega_{\text{max}}$ is the maximum frequency available, $\sigma$ is the width of the Gaussian used to smooth $\tilde{C}(\omega)$, and $T$ is the total time. The bias for random fields is

$$E_{\text{bias}} = \left( \frac{M(M - 1)}{2} + \frac{3M}{8} \right) \frac{\sqrt{\pi}}{T\sigma_\omega} \prod_{i=1}^{d} L_i \sigma_{q_i}$$

$$\dot{E}_{\text{bias}} = \left( \frac{M(M - 1)}{2} + \frac{3M}{8} \right) \frac{\omega_{\text{max}}}{T\sigma_\omega \sqrt{\pi}} \prod_{i=1}^{d} \frac{q_{i,\text{max}}}{L_i \sigma_{q_i} \sqrt{\pi}}$$

(15)

(16)

where $L_i$ is the length, $q_{i,\text{max}}$ is the maximum wavenumber, and $\sigma_{q_i}$ is the width of the Gaussian used to smooth $\tilde{C}(\mathbf{q}, \omega)$ in the $i^{th}$ spatial dimension.

C. Simulations

To simulate the driven Gaussian fields, Eq. 5, we nondimensionalize the system of equations using a time scale $\tau = 1/(Dr)$ and length scale $\lambda = 1/\sqrt{r}$. We use an Euler-Maruyama algorithm to simulate the dynamics of the two fields on a periodic, 1 dimensional lattice.

We simulate Eq. 8 using Gillespie’s algorithm [42] to create a stochastic trajectory through the $(X,Y)$ phase plane with a well-mixed volume of $V = 100$. We calculate the true $\dot{S}$ of any specific trajectory $\mathbf{z} = \{m_i|i = 1, \ldots, N\}$ as follows. For each state $m'$, there exists a probability per unit time of transitioning to a new state $m$ via a chemical reaction $\mu$, denoted by $W_{m,m'}^{(\mu)}$. At steady state, the true entropy produced is [34]

$$\Delta S_{\text{true}}[\mathbf{z}] = \sum_{j=1}^{N} \ln \frac{W_{m_j,m_{j-1}}^{(\mu_j)}}{W_{m_{j-1},m_j}^{(\mu_j)}}$$

(17)

Note that $\Delta S_{\text{true}}$ is now itself a random variable that depends on the specific trajectory. We estimate $\langle \dot{S}_{\text{true}} \rangle$ by fitting a line to an ensemble average of $\Delta S_{\text{true}}$ (Supplementary Figure 1), and compare that to $\hat{S}$. We calculate $\dot{S}_{\text{blind}}$ by summing over all the reactions that can give rise to the observed transition at every time point along the trajectory.
To simulate the reaction-diffusion Brusselator, Eq. 10, we take a compartment-based approach [43] where we treat each chemical species in each compartment as a separate species, and treat diffusion events as additional chemical reaction pathways. We nondimensionalize time by $\tau = 1/k_1^+$ and use a Gillespie algorithm to simulate all reactions on a 1 dimensional periodic lattice with $L$ sites.

See Supplementary Materials for all simulation parameters used in each figure.

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Dissipative signatures of dynamical phases and transitions

Supplementary Materials

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I. SUPPLEMENTARY FIGURES

Supplementary Figure 1. Fit to (blinded) entropy produced for Brusselator. Light gray lines show the amount of entropy produce as a function of simulation time for \( N = 50 \) simulations at \( \Delta \mu = 4.5 \). Each simulation starts at a random initial condition and rapidly approaches the steady state value for \((X,Y)\). This transient trajectory results in the large variation in initial entropy production which depends on how far the system begins from \((X_{ss},Y_{ss})\). Once the system reaches its steady state, the rate of entropy production approaches a steady value. The average of \( \Delta S \) is taken across all trajectories, and a linear fit to the second half of the resulting mean gives us our value of \( \dot{S}_{\text{blind}} \) given in Fig. 3b. The same method is used to calculate \( \dot{S}_{\text{true}} \) as well as \( \dot{S}_{\text{blind}} \) and \( \dot{S}_{\text{true}} \) for the reaction-diffusion Brusselator model in Fig. 4.

Supplementary Figure 2. (left) Comparison of \( \dot{S}_{\hat{\mu}} \) and \( \dot{S}_{\nu^{2}} \), showing qualitatively similar results, indicating that \( \dot{S}_{\hat{\mu}} \) contains the same information as a Fokker-Planck description of the system. (right) Same plot on a log-log scale. The red dashed lines have a slope of 2, reflecting the fact that the entropy production rate near equilibrium is quadratic in the driving force \( \Delta \mu \). While \( \dot{S}_{\hat{\mu}} \) is quadratic, \( \dot{S}_{\nu^{2}} \) is not, showing us that \( \dot{S}_{\hat{\mu}} \) is a more accurate measure of \( \dot{S}_{\text{true}} \) near equilibrium.
Supplementary Figure 3. Brownian particle. (a) Sample trajectory for simulation of driven Brownian particle in 2 dimensions with $\alpha = 2$ in nondimensionalized units shown in gray with end of trajectory shown with white circle. The heatmap gives the empirical steady-state probability distribution function of particle positions and the red arrows indicate the underlying force field $F(x)$. (b) $E$ for $\alpha = [1, 3, 9]$ measured from simulations and calculated from Eq. 74, shown in solid and dashed lines respectively. $E$ is symmetric in $\omega$, so only the positive axis is shown. (c) EPR for $\alpha = \{0, 1, \ldots, 10\}$, smoothed by a Gaussian with $\sigma_\omega = 2.1$. Mean ± standard deviation of $\dot{S}$ over $N = 64$ simulations shown with black dots and shaded area. Red line shows non-dimensionalized theoretical value of $\dot{S}$. See Supplementary Materials for all simulation parameters.

Supplementary Figure 4. $\dot{S}$ for driven Brownian particle simulation in $d = [2, 3, 4]$. Left plot is the same as in the main text, and the other two plots show equivalent information, with all other parameters the same as in Fig. 1b.

II. DERIVATION OF THE ENTROPY PRODUCTION FACTOR

Consider a system described by a set of real random scalar variables tracing some path through phase space, $z = \{X_i(t)\}$. Assuming the variables to be Gaussian distributed with real space covariance function $\langle X^i(t)X^j(t') \rangle = C^{ij}(t - t')$, the probability of observing a particular path is given in frequency space as

$$ P[z] = \frac{1}{Z} \exp \left[ -\frac{1}{2} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} C_{ij}^{-1}(\omega)x^i(-\omega)x^j(\omega) \right] $$

where $Z$ is the partition function and $C^{ij}(\omega) = \mathcal{F}[C^{ij}(t - t')]$ with $C_{ij}^{-1} \equiv (C^{-1})_{ij}$. The reverse path is given by Eq. 1 with $\omega \rightarrow -\omega$ in the argument of $C_{ij}^{-1}$.

$$ P[\tilde{z}] = \frac{1}{\tilde{Z}} \exp \left[ -\frac{1}{2} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} C_{ij}^{-1}(-\omega)x^i(-\omega)x^j(\omega) \right] . $$

(2)
Supplementary Figure 5. Effects of smoothing on $\dot{S}$. (a) Real part of $C_{xx}(\omega) = \langle x(\omega)x^*(\omega) \rangle$ and $C_{xy}(\omega) = \langle x(\omega)y^*(\omega) \rangle$ for 2d driven Brownian particle simulations. Raw covariance functions shown in grey, with the results of smoothing with increasingly wide Gaussians shown in color, from purple to yellow for narrowest to widest. (b) Similar to (a), but shows the imaginary part of $C_{xx}$ and $C_{xy}$. Note that $C$ is Hermitian, $C^\dagger = C$, so the other elements $C$ contain identical information. (c) Values of $\dot{S}$ at $\alpha = 2$ as a function of both amount of data and smoothing for $d = [2, 3, 4]$ simulations of a driven Brownian particle.

To make the following calculations easier, we consider a discrete case for a finite time series of length $T = Ndt$ with sampling rate $dt$. In this case, Eq. 1 is written as

$$P[z] = \frac{1}{Z} \prod_{n=1}^{T/dt} \exp \left[ -\frac{1}{2T} [C^{-1}(\omega_n)]_{ij} x_i(-\omega_n)x_j^*(\omega_n) \right]$$

where $\omega_n = 2\pi n/T$. Eq. 2 is written similarly. We then have

$$\ln \left( \frac{P}{\tilde{P}} \right) = \ln \left( \frac{\tilde{Z}}{Z} \right) + \frac{1}{2T} \sum_{n=1}^{T/dt} [C^{-1}(-\omega_n) - C^{-1}(\omega_n)]_{ij} x_i(-\omega_n)x_j^*(\omega_n)$$

Using the fact that, for a finite signal of length $T$, $\langle x_i(\omega_n)x_j(-\omega_m) \rangle = T \delta_{nm} C_{ij}(\omega_n)$, the KL-divergence is then

$$D_{KL}(P[z] \mid \tilde{P}[\tilde{z}]) = \left\langle \ln \left( \frac{P}{\tilde{P}} \right) \right\rangle$$

$$= \sum_{z} P \ln \left( \frac{P}{\tilde{P}} \right)$$

$$= \ln \left( \frac{\tilde{Z}}{Z} \right) + \frac{1}{2T} \sum_{n} [C^{-1}(-\omega_n) - C^{-1}(\omega_n)]_{ij} C^{ji}(\omega_n)$$

where $\sum_{z}$ is a sum over all possible paths. The entropy production rate is then given by

$$\dot{S} = \lim_{T \to \infty} \frac{1}{2T} \sum_{n} [C^{-1}(-\omega_n) - C^{-1}(\omega_n)]_{ij} C^{ji}(\omega_n) = \lim_{T \to \infty} \frac{1}{T} \sum_{n} \mathcal{E}(\omega_n),$$

where we have introduced the entropy production factor, $\mathcal{E}$, and dropped the ratio of the partition functions as they will contribute 0 to the EPR when multiplied by $1/T$ and taking the $T \to \infty$ limit. In the limit taken, the sum becomes an integral, $\sum_{n} \to T/2\pi \int d\omega$, which brings us to our first main result

$$\dot{S} = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \mathcal{E}(\omega)$$
\( E \) can also be rewritten as a 
\[
E(\omega) = \text{Tr}\{C(\omega)\left[ C^{-1}(-\omega) - C^{-1}(\omega) \right]\} / 2.
\]

In the same spirit as above, we derive a similar expression for a set of real-valued random fields, \( \eta(r, t) = \{\phi^i(r, t) | r \in \mathbb{R}^d \} \). The probability of following a path is given by

\[
P[\eta] = \frac{1}{Z} \exp \left[ -\frac{1}{2} \int \frac{d\omega}{2\pi} \int \frac{d^d q}{(2\pi)^d} C_{ij}(q, \omega) \phi^i(-q, -\omega) \phi^j(q, \omega) \right],
\]

where the covariance function is defined as \( \langle \phi^i(q, \omega) \phi^j(q', \omega') \rangle = C^{ij}(q, \omega) \delta^d(q + q') \delta(\omega + \omega') \). Under time reversal, \( q \) is invariant but \( \omega \) switches sign. Thus, \( P[\eta] \) is given by

\[
P[\eta] = \frac{1}{Z} \exp \left[ -\frac{1}{2} \int \frac{d\omega}{2\pi} \int \frac{d^d q}{(2\pi)^d} C_{ij}(q, \omega) \phi^i(-q, -\omega) \phi^j(q, \omega) \right].
\]

We assume that the field is sampled in time with resolution \( dt \) for a time \( T \) (i.e. \( \Delta \omega = 2\pi/T \)) and each dimension of space is sampled with resolution \( dx_i \), for a length \( L_i \) (i.e. \( \Delta k_i = 2\pi/L_i \)), giving the discretized path probability functional

\[
P[\eta] = \frac{1}{Z} \prod_{n=0}^{T/dt} \prod_{m_1=0}^{L_1/dx_1} \cdots \prod_{m_d=0}^{L_d/dx_d} \exp \left[ -\frac{1}{2VT} C_{ij}^{-1}(q_{m, \omega_{n}}) \phi^i(-q_{m}, -\omega_{n}) \phi^j(q_{m}, \omega_{n}) \right],
\]

where \( q_m = (k_{1m_1}, k_{2m_2}, \ldots, k_{d_m}) \) and \( V \) is the total volume. A similar expression exists for \( P[\eta] \). From these expressions, we have (ignoring the partition functions that will add zero once we take the \( T \rightarrow \infty \) limit)

\[
\ln \left( \frac{P}{\bar{P}} \right) = -\frac{1}{2VT} \sum_{n=0}^{T/dt} \sum_{m_1=0}^{L_1/dx_1} \cdots \sum_{m_d=0}^{L_d/dx_d} \left[ C_{ij}^{-1}(q_{m, \omega_{n}}) - C_{ij}^{-1}(q_{m}, \omega_{n}) \right] \delta^d(-q_{m}, -\omega_{n}) \phi^j(q_{m}, \omega_{n}).
\]

Taking the average with respect to \( P \) and noting that \( \langle \phi^i(q_{m}, \omega_{n}) \phi^j(-q_{m'}, -\omega_{n'}) \rangle = TV \delta_{mm'} \delta_{nn'} \delta^d(q_{m}, \omega_{n}) \) for finite signals, we have

\[
D_{KL}(P[\eta] \mid P[\bar{\eta}]) = -\frac{1}{2} \sum_{n=0}^{T/dt} \sum_{m_1=0}^{L_1/dx_1} \cdots \sum_{m_d=0}^{L_d/dx_d} \left[ C_{ij}^{-1}(q_{m, \omega_{n}}) - C_{ij}^{-1}(q_{m}, \omega_{n}) \right] \delta^d(-q_{m}, -\omega_{n}) C^{ij}(q_{m}, \omega_{n}).
\]

This gives an entropy production rate of

\[
\dot{S} = \lim_{T \rightarrow \infty} \frac{1}{2T} \sum_{n=0}^{T/dt} \sum_{m_1=0}^{L_1/dx_1} \cdots \sum_{m_d=0}^{L_d/dx_d} E(q_{m}, \omega_{n}),
\]

again introducing the EPF for fields, \( E(q, \omega) \). Passing to the continuum limit, we have

\[
\dot{S} = V \int \frac{d\omega}{2\pi} \int \frac{d^d q}{(2\pi)^d} E(q, \omega),
\]

The EPR density is given by \( \dot{s} = \dot{S} / V \). As with the case of random variables, \( E \) can be rewritten as a trace, 
\( E(q, \omega) = \text{Tr}\{C^{-1}(q, -\omega) - C^{-1}(q, \omega)\} C(q, \omega) / 2 \).

### A. Numerical calculations

#### 1. Partition functions

While we were able to drop the partition functions in the analytic calculations above after taking the \( T \rightarrow \infty \) limit and assuming translation and rotation invariance, the case of finite, discrete data requires greater care due to the presence of noise. The partition function for Eq. 10 is given by

\[
Z = \exp \left( \frac{VT}{2} \int \frac{d\omega}{2\pi} \frac{d^d q}{2\pi} \ln \left| \text{det} C(q, \omega) \right| \right).
\]
and the EPR density is given by
\[
\dot{s} = \frac{1}{2} \iint \frac{d^d \mathbf{q}}{(2\pi)^d} \frac{d\omega}{2\pi} \left[ \ln \left( \frac{\det C(q, -\omega)}{\det C(q, \omega)} \right) + \left[ C^{-1}(q, -\omega) - C^{-1}(q, \omega) \right]_{ij} C^{ji}(q, \omega) \right].
\] (18)

In discrete form,
\[
\dot{s} = \frac{1}{2TV} \sum_{m,n} \left[ \ln \left( \frac{\det C(q_m, -\omega_n)}{\det C(q_m, \omega_n)} \right) + \left[ C^{-1}(q_m, -\omega_n) - C^{-1}(q_m, \omega_n) \right]_{ij} C^{ji}(q_m, \omega_n) \right].
\] (19)

For random variables, the equivalent of Eq. 17 has an integral over only \(\omega\) in the argument of the exponential resulting in the same value of \(Z\) since the integral is over all \(\omega\). This gives \(\ln(Z/Z) = 0\), allowing one to drop the partition functions even for finite trajectories. However, for finite trajectories of fields, the partition functions are required to maintain the positivity of the relative entropy used to define \(\dot{s}\).

2. First moment properties

We now turn to the problem of estimating the bias in our measured entropy production rate. For this, we assume that we have an equilibrium process and calculate what the average measured entropy production rate is, representing the systematic overestimation of our estimator. We work in coordinates where the covariance matrix is the identity, \(C^{\mu\nu} = \delta^{\mu\nu}\). Due to a combination of measurement errors and only having a finite time series, we will measure a matrix that deviates from the identity by
\[
\tilde{C}^{\mu\nu} = \delta^{\mu\nu} + \tilde{R}^{\mu\nu} + i\tilde{A}^{\mu\nu},
\] (20)
where \(\tilde{R}^{\mu\nu}(\omega)\) and \(\tilde{A}^{\mu\nu}(\omega)\) are elements of a symmetric and anti-symmetric \(D \times D\) matrix, respectively, each assumed to be much smaller than 1. The anti-symmetric contribution must be purely imaginary because \(C^{\mu\nu}\) is a Hermitian matrix by definition. Further, we have \(R^{\mu\nu}(-\omega) = R^{\mu\nu}(\omega)\) and \(A^{\mu\nu}(-\omega) = -A^{\mu\nu}(\omega)\). For notational simplicity, we define \(\tilde{M}^{\mu\nu} = \delta^{\mu\nu} + \tilde{R}^{\mu\nu}\) and therefore \(\tilde{C} = \tilde{M} + i\tilde{A}\).

To calculate the EPR, we need to calculate the EPF, \(E = \text{Tr}\{C(\omega) \left[ C^{-1}(-\omega) - C^{-1}(\omega) \right]\}\). We approximate \(\tilde{C}^{-1}\) as
\[
\tilde{C}^{-1} = (\tilde{M} + i\tilde{A})^{-1} \approx \tilde{M}^{-1} - i\tilde{M}^{-1}\tilde{A}\tilde{M}^{-1}.
\] (21)

Then, \(C^{-1}(-\omega) - C^{-1}(\omega) = 2i\tilde{M}^{-1}\tilde{A}\tilde{M}^{-1}\). Multiplying by \(\tilde{C}\),
\[
C(\omega) \left[ C^{-1}(-\omega) - C^{-1}(\omega) \right] = 2i\tilde{M}^{-1}\tilde{A}\tilde{M}^{-1}\tilde{M} - 2\tilde{M}^{-1}\tilde{A}\tilde{M}^{-1}\tilde{A}
\] (22)

Taking the trace of Eq. 22, the first term is an asymmetric matrix with zero trace. By writing \(\tilde{M} = \mathbb{I} + \tilde{R}\), we approximate \(\tilde{M}^{-1} \approx \mathbb{I} - \tilde{R}\), and the second term is approximately as \(\tilde{A}^2 + \mathcal{O}(\tilde{A}^2\tilde{R} + \tilde{A}\tilde{R}\tilde{A})\). Thus, to lowest order we have
\[
E = \text{Tr}\{C(\omega) \left[ C^{-1}(-\omega) - C^{-1}(\omega) \right]\} \approx -\text{Tr}\left(\tilde{A}^2\right) = 2 \sum_{\mu > \nu} (\tilde{A}^{\mu\nu})^2.
\] (23)

Assuming each element of \(\tilde{A}\) is an iid random variable, we can write the average EPF measured at equilibrium as
\[
\langle E \rangle_{eq} = 2 \frac{M(M-1)}{2} \langle (\tilde{A}^{\mu\nu})^2 \rangle = \frac{M(M-1)}{2}.
\] (24)

We calculated \(\langle (\tilde{A}^{\mu\nu})^2 \rangle\) as follows:
\[
\langle (\tilde{A}^{\mu\nu})^2 \rangle = \langle \text{Im}(x^{\mu\nu}x^{\mu\nu}) \rangle^2 = \langle \text{Re}(x^{\mu\nu})\text{Im}(x^{\mu\nu}) + \text{Im}(x^{\mu\nu})\text{Re}(x^{\mu\nu}) \rangle^2
\] (25)
\[
= \langle \text{Re}(x^{\mu\nu})\text{Im}(x^{\mu\nu}) \rangle^2 + \langle \text{Im}(x^{\mu\nu})\text{Re}(x^{\mu\nu}) \rangle^2 + \text{cross-terms}
\] (26)
\[
= 1/2
\] (27)
The cross-terms average to zero because, in our choice of coordinate system, \(|x^\mu|^2 = 1\), so the real and imaginary parts of \(x^\mu\) are equally distributed along the unit circle. In addition, \(x^\mu\) and \(x^\nu\) should be uncorrelated (recall that we are working at equilibrium). The first and second term each have both a real and an imaginary part squared, each of which is always positive and on average equal to 1/2. Thus, each term is 1/4 and adds to 1/2.

To estimate \(\dot{S}\), we smooth \(\tilde{E}\) and integrate over all frequencies. We calculate \(\langle (\dot{A}^{\mu\nu})^2 \rangle\) as

\[
\langle (\dot{A}^{\mu\nu})^2 \rangle = \left( \frac{\sum \Delta \omega \exp[-(\omega_i - \omega_n)^2/2\sigma^2]}{\sqrt{2\pi}\sigma^2} \tilde{A}_{\mu\nu}(\omega_i) \right)^2
\]

\[
\langle (\dot{A}^{\mu\nu})^2 \rangle = \sum_{\omega_i} (\Delta \omega)^2 \exp[-(\omega_i - \omega_n)^2/2\sigma^2] \langle (\dot{A}^{\mu\nu})^2(\omega_i) \rangle
\]

\[
\approx \frac{\Delta \omega}{4\pi\sigma^2} \int d\omega \exp[-\omega^2/\sigma^2] = \frac{\Delta \omega}{4\pi\sigma^2} \sqrt{\frac{\pi}{2T\sigma}}
\]

We used \(\langle \tilde{A}^{\mu\nu}(\omega_i) \tilde{A}^{\mu\nu}(\omega_j) \rangle = \langle (\dot{A}^{\mu\nu})^2(\omega_i) \rangle \delta_{ij}\) in the second line, passed to an integral using one of the integration measures \(\Delta \omega\) in the third line, and substituted \(\Delta \omega = 2\pi/T\) in the fourth line. Finally, we arrive at

\[
\dot{S}_{eq} = \frac{2}{N} \frac{M(M - 1)}{2} \int_{-\omega_{max}}^{\omega_{max}} \frac{d\omega}{2\pi} \langle (\dot{A}^{\mu\nu})^2 \rangle = \frac{M(M - 1)}{2} \frac{\omega_{max}}{T\sigma\sqrt{\pi}}
\]

If we also average the covariance functions over \(N\) independent trajectories with the same dynamics, this bias is further reduced, leaving us with our final estimate of the bias in our entropy production rate estimator

\[
\dot{S}_{eq} = \frac{1}{N} \frac{M(M - 1)}{2} \frac{\omega_{max}/\sigma}{T\sqrt{\pi}}
\]

Following the same line of reasoning for a set of \(M\) fields in \(d + 1\) dimensions, a similar expression can be derived. We again write \(C^{\mu\nu}(q, \omega) = 1 + R^{\mu\nu}(q, \omega) + A^{\mu\nu}(q, \omega)\). Extra care must be taken in the field case because S.Eq. 16 does not have the same symmetries as S.Eq. 9. Specifically, while \(R^{\mu\nu}(-q, -\omega) = R^{\mu\nu}(q, \omega)\) and \(A^{\mu\nu}(-q, -\omega) = -A^{\mu\nu}(q, -\omega)\), nothing can be said \textit{a priori} about \(R(q, -\omega)\) or \(A(q, -\omega)\).

In order to calculate the bias, we will calculate the mean of \(\mathcal{E} = \text{Tr}\{\left[ \mathcal{C}^{-1}(q, -\omega) - \mathcal{C}^{-1}(q, \omega) \right] \mathcal{C}(q, \omega) \}\) The calculation is tedious, so we only report the result here:

\[
\langle \mathcal{E} \rangle_{eq} = \langle \text{Tr}\left[ \mathcal{R}^2 - \mathcal{A}^2 \right]\rangle
\]

\[
= M(M - 1) \left( \langle (\mathcal{R}^{\mu\nu})^2 \rangle + \langle (\mathcal{A}^{\mu\nu})^2 \rangle \right) + M \langle (\mathcal{R}^{\mu\nu})^2 \rangle
\]

\[
= M(M - 1) + 3M/4
\]

We arrived at this by using the fact that \(\text{Tr}\left( \mathcal{A}^2 \right) = \sum_{\mu \neq \nu} \langle A^{\mu\nu} \rangle^2\) and \(\text{Tr}\left( \mathcal{R}^2 \right) = \sum_{\mu \neq \nu} \langle R^{\mu\nu} \rangle^2 + \sum_{\mu \neq \nu} \langle R^{\mu\nu} \rangle^2\) for an asymmetric and symmetric matrix, respectively, in addition to the assumption that every matrix element is an i.i.d. random variable. As before, \(\langle (\mathcal{A}^{\mu\nu})^2 \rangle = 1/2 = \langle (\mathcal{R}^{\mu\nu})^2 \rangle\). Now turning to the diagonal elements of \(\mathcal{R}\),

\[
\langle (\mathcal{R}^{\mu\nu})^2 \rangle = \langle [1 - \text{Re}(\phi^\mu \phi^\nu)]^2 \rangle
\]

\[
= 1 + \langle \text{Re}(\phi^\mu)^2 \text{Im}(\phi^\nu)^2 \rangle + \langle \text{Re}(\phi^\nu)^4 \rangle + \langle \text{Im}(\phi^\mu)^4 \rangle - 2\langle \text{Re}(\phi^\mu)^2 \rangle^2 \text{Im}(\phi^\nu)^2 \rangle
\]

\[
= 3/4
\]

where we used \(\langle x^4 \rangle = 3\langle x^2 \rangle^2\) for Gaussian variables and \(\langle \text{Re}(\phi^\mu)^2 \rangle = \langle \text{Im}(\phi^\mu)^2 \rangle = 1/2\) in our choice of coordinate system.

Assuming the signals have a total length in time of \(T\) and a total length in each spatial dimension of \(L_i\), we smooth the spatiotemporal covariance function with a multivariate Gaussian of width \(\sigma_\omega\) in the temporal dimension, and \(\sigma_{k_i}\) in each of the spatial dimensions, giving a factor of \(\omega_{max}/\sigma_\omega T\sqrt{\pi}\) for the temporal dimension and \(q_{i,\text{max}}/\sigma_{q_i} L_i\sqrt{\pi}\) for each spatial dimension. Putting all these results together, we have

\[
\dot{s}_{eq} = \frac{1}{N} \left( \frac{M(M - 1)}{2} + \frac{3M}{8} \right) \frac{\omega_{max}}{T\sigma_\omega \sqrt{\pi}} \prod_{i=1}^{d} \frac{q_{i,\text{max}}}{L_i \sigma_{q_i} \sqrt{\pi}}
\]
III. EPR OF COUPLED GAUSSIAN FIELDS

Consider the coupled equations of motion for the scalar fields $\phi$ and $\psi$ in $d + 1$ dimensions.

\[
\partial_t \phi(x, t) = -D(r - \nabla^2)\phi - \alpha \psi + \sqrt{2D} \xi_\phi \tag{42}
\]
\[
\partial_t \psi(x, t) = -D(r - \nabla^2)\psi + \alpha \phi + \sqrt{2D} \xi_\psi. \tag{43}
\]

with $\langle \xi^i(x, t)\xi^j(x', t') \rangle = \delta^{ij}\delta(t - t')\delta^d(x - x')$. This is a Gaussian model with free energy

\[
F = \int d^d x \left[ \frac{r}{2} (\phi^2 + \psi^2) + \frac{1}{2} (|\nabla \phi|^2 + |\nabla \psi|^2) \right]. \tag{44}
\]

The interaction term cannot be written as a gradient of an energy, so we have

\[
\partial_t \phi(x, t) = -D \frac{\delta F}{\delta \phi} - \alpha \psi + \sqrt{2D} \xi_\phi \tag{45}
\]
\[
\partial_t \psi(x, t) = -D \frac{\delta F}{\delta \psi} + \alpha \phi + \sqrt{2D} \xi_\psi. \tag{46}
\]

Written in vector notation,

\[
\partial_t \mathbf{\eta}(x, t) = A \mathbf{\eta}(x, t) + \sqrt{2D} \mathbf{\xi}(x, t); \quad B(x) = \begin{pmatrix} -D(r - \nabla^2) & -\alpha \\ \alpha & -D(r - \nabla^2) \end{pmatrix}; \quad \mathbf{\eta}(x, t) = \begin{pmatrix} \phi(x, t) \\ \psi(x, t) \end{pmatrix}. \tag{47}
\]

To get the cross-spectral density, we rewrite S.Eq. 47 as an Ito stochastic differential equation:

\[
d\mathbf{\eta} = B \mathbf{\eta} dt + \Xi d\mathbf{W}, \tag{48}
\]

where $\mathbf{W}(x, t)$ is a multidimensional Wiener process in space and time with strength $\Xi^{ij} = \sqrt{2D} \delta^{ij}$. The eigenvalues of $A$ have negative real parts, so a stationary solution exists. The cross-spectral density is [1]

\[
C(q, \omega) = (B(q) - i\omega I)^{-1} \Xi^{T} (B(q) + i\omega I)^{-T}. \tag{49}
\]

Noting $\Xi^{T} = 2D I$, we have

\[
C(\omega) = \frac{2D}{|D(r + q^2)|^2 + i\omega^2 + \alpha^2} \left( \frac{(D(r + q^2))^2 + \alpha^2 + \omega^2}{-i2\alpha \omega} \right) \left( \frac{i2\alpha \omega}{[D(r + q^2)]^2 + \alpha^2 + \omega^2} \right). \tag{50}
\]

The inverse is given by

\[
C^{-1}(\omega) = \frac{1}{2} \begin{pmatrix} (D(r + q^2))^2 + \alpha^2 + \omega^2 & -i2\alpha \omega \\ i2\alpha \omega & [D(r + q^2)]^2 + \alpha^2 + \omega^2 \end{pmatrix}. \tag{51}
\]

Finally, using S.Eq. 16, we have

\[
\hat{S} = V \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{d\mathbf{q}}{2\pi} \frac{8\alpha^2 \omega^2}{|([D(r + q^2)]^2 + i\omega^2 + \alpha^2)|^2} = V \frac{\alpha^2}{D\sqrt{\pi}}. \tag{52}
\]

Rearranging the denominator of the integrand of above gives $E^{DG}$ given in the main text.

We can alternatively calculate the entropy production rate by using the Onsager-Machlup functional [2] for the path probability functional $P[\mathbf{\eta}]$ in

\[
\hat{S} = \lim_{T \to \infty} \frac{1}{T} \left\langle \ln \frac{P[\mathbf{\eta}]}{P[\mathbf{\eta}]} \right\rangle. \tag{53}
\]

Writing it as a path $P[\mathbf{\eta}] \propto \exp(-A)$, where $A$ is the action, this becomes

\[
\hat{S} = \lim_{T \to \infty} \frac{1}{T} \left\langle \tilde{A} - A \right\rangle, \tag{54}
\]
where $\tilde{A}$ is the action under time-reversal. To calculate $A$, we use standard path integral techniques, i.e. the Martin-Siggia-Rose formalism [3]. The idea is to try and find the expectation of some observable, $O$, over noise realizations.

$$\langle O|\xi\rangle = \int D[\xi] O[\xi] P[\xi].$$

(55)

Since the noise is Gaussian, we have

$$P[\xi] \propto \exp\left(\frac{1}{4D} \int d^4x dt \, \xi^2\right)$$

(56)

(we use Einstein notation throughout). We then insert the most complicated expression for 1 ever written. Using the integral representation of the functional delta function, $\delta[f(x)] = \int D[i\bar{f}] \exp[-\int dx \bar{f}(x)f(x)]$, we write

$$1 = \int \prod_j D[\eta^j] \delta(\partial_t \eta^j - B_k^j \eta^k - \xi^j)$$

$$= \int \prod_j D[\eta^j] D[i\bar{\eta}_j] \exp - \int d^4x dt \left[ \bar{\eta}_j \left( \partial_t \eta^j - B_k^j \eta^k - \xi^j \right) \right]$$

(57)

(58)

to get

$$\langle O|\xi\rangle = \int D[\xi] \prod_j D[\eta^j] D[i\bar{\eta}_j] O[\eta] \exp \left[ \frac{1}{4} \int d^4x dt \left( \xi^i \Xi_{ij}^{-1} \xi^j - 4\bar{\eta}_j \xi^j \right) - \bar{\eta}_j \left( \partial_t \eta^j - B_k^j \eta^k \right) \right].$$

(59)

Completing the square in $\xi$ and doing the Gaussian integrals, we get

$$\langle O|\xi\rangle = \int \prod_j D[\eta^j] D[i\bar{\eta}_j] O[\eta] \times \exp \left\{ - \int d^4x dt \left[ \bar{\eta}_j \left( \partial_t \eta^j - B_k^j \eta^k \right) - \bar{\eta}_j \Xi_{jk} \bar{\eta}_k \right] \right\}. $$

(60)

Doing the integrals over the response fields $\bar{\eta}_j$ we are left with

$$\langle O|\eta\rangle = \int \prod_j D[\eta^j] O[\eta] \exp(-A[\eta])$$

(61)

where $A$ is the Onsager-Machlup functional

$$A = -\frac{1}{4D} \int d^4x dt \, \left( \partial_t \eta^j - B_k^j \eta^k \right)^2$$

(62)

Noting that the only time asymmetric part of the action is $\partial_t \eta$, we can write

$$A = -\frac{1}{4D} \int d^4x dt \, \left( \partial_t \phi + D \frac{\delta F}{\delta \phi} + \alpha \psi \right)^2 + \left( \partial_t \psi + D \frac{\delta F}{\delta \psi} - \alpha \phi \right)^2$$

(63)

$$\tilde{A} = -\frac{1}{4D} \int d^4x dt \, \left( \partial_t \phi - D \frac{\delta F}{\delta \phi} - \alpha \psi \right)^2 + \left( \partial_t \psi - D \frac{\delta F}{\delta \psi} + \alpha \phi \right)^2$$

(64)

Taking the difference $\tilde{A} - A$, and noting that $(a + b)^2 - (a - b)^2 = 4ab$, we have

$$\tilde{A} - A = -\frac{1}{D} \int d^4x dt \, \partial_t \phi \left( -D \frac{\delta F}{\delta \phi} - \alpha \psi \right) + \partial_t \psi \left( -D \frac{\delta F}{\delta \psi} + \alpha \phi \right)$$

(65)

In the Stratonovich convention, $dF/dt = \partial_t \phi \left( \delta F/\delta \phi \right) + \partial_t \psi \left( \delta F/\delta \psi \right)$, which will turn into a constant difference in free energies upon taking the time integral, allowing us. This constant value will tend to zero as the limit $T \to \infty$ is taken. Further, there is a time-symmetric portion of the action that is being omitted due to the Jacobian factor in switching from an integral in $\xi$ to $\eta$ that also arises due to the Stratonovich discretization used throughout this article.

We find the entropy production rate to be

$$\dot{S} = \lim_{T \to \infty} \frac{\alpha}{DT} \int d^4x dt \, \left( \dot{\phi} - \dot{\psi} \right)$$

(66)
Plugging in the equations of motion, we find
\[ \langle \dot{\psi} \phi \rangle = \langle -D \delta F / \delta \psi \phi - \alpha \phi^2 + \xi \psi \phi \rangle = \langle -D \left[ (r - \nabla^2) \psi \right] \phi - \alpha \phi^2 + \xi \psi \phi \rangle \] (67)
\[ \langle \dot{\phi} \psi \rangle = \langle -D \delta F / \delta \phi \psi + \alpha \psi^2 + \xi \phi \psi \rangle = \langle -D \left[ (r - \nabla^2) \phi \right] \psi + \alpha \psi^2 + \xi \phi \psi \rangle \] (68)

Some care must be taken in evaluating the terms linear in the noise. If the Ito convention had been used, they would be trivially zero, but that is not the general case in the Stratonovich convention. However, as each field is multiplied by the opposite component of the noise, one can show that they indeed identically equal 0. Putting everything together, we have
\[ -Dr \psi \phi + D \left( r - \nabla^2 \right) \psi \phi + \alpha \phi^2 + \xi \psi \phi \] (69)

The two Laplacian terms will cancel under one integration by parts each, leaving us with
\[ \dot{S} = \frac{\alpha^2}{D} \int d^d x \left( \phi^2 + \psi^2 \right) \] (70)

where we have replaced the time average with an ensemble average, assuming ergodicity. Assuming the system to be in the steady state, we integrate over the equal-time (i.e. \( \omega = 0 \)) power spectrum of \( \phi \) and \( \psi \)
\[ \langle \phi(k) \phi(-k) \rangle = \int \frac{dk}{2\pi k^2 + r} = \frac{1}{2\sqrt{r}}. \] (71)

Using this expression for both \( \phi \) and \( \psi \) in the equation for \( \dot{S} \) above, we have
\[ \dot{S} = \frac{\alpha^2}{D\sqrt{r}} V, \] (72)

where \( V \) is the total volume of the space, and \( \dot{s} = \dot{S}/V \).

**IV. DRIVEN BROWNIAN PARTICLE**

Here, we consider the particle version of the dynamics given by the Gaussian fields above. We consider an overdamped Brownian particle in a 2-dimensional harmonic trap with stiffness \( r \) subject to a non-conservative, rotational force. This is the particle version of the driven Gaussian fields considered above. The dynamics obey the Langevin equation
\[ \dot{x} = Ax + \sqrt{2} \xi; \quad A = \begin{pmatrix} -r & -\alpha \\ \alpha & -r \end{pmatrix}, \] (73)

where \( \xi \) is Gaussian white noise, \( \langle \xi_i(t) \xi_j(t') \rangle = \delta_{ij} \delta(t - t') \). These linear dynamics exactly satisfy Eq. 1 [1]. Using S.Eq. 9 with the analytically calculated covariance functions, we find the exact EPF, integrating to yield the EPR, in agreement with results from stochastic thermodynamics [4]
\[ \mathcal{E}^{DBP} = \frac{8\alpha^2 \omega^2}{(\omega^2 - \omega_0^2)^2 + (2r\omega)^2}; \quad \dot{S}^{DBP} = \frac{2\alpha^2}{r} \] (74)

The covariance functions are calculated in the same way we calculated the covariance functions for the driven Gaussian fields, simply replacing \( D(r + q^2) \rightarrow r \). For the solution using stochastic thermodynamics, we write the Fokker-Planck equation associated with S.Eq. 73 and solving for the steady state probability density, \( p_{ss} \) as well as the steady state current, \( j_{ss} \), given by
\[ p_{ss}(x) = \frac{r}{2\pi} e^{-rx^2/2}; \quad j_{ss}(x) = \alpha x p_{ss}. \] (75)

We can then calculate \( \dot{S} \) as [4]
\[ \dot{S} = \int dx \frac{j_{ss}^2}{p_{ss}} = \frac{2\alpha^2}{r} \] (76)
Similarly to $\mathcal{E}_{DGF}$, $\mathcal{E}_{DBP}$ is peaked at $\omega_0 = (r^2 + \alpha^2)^{1/2}$ and decays as $\omega^2$ for large $\omega$. While multiple combinations of $\alpha$ and $r$ can give the same value for $\hat{S}$, $\mathcal{E}$ distinguishes between equally dissipative trajectories in the shape and location of its peaks, giving information about the form of the underlying dynamics while retaining the same total EPR.

To test whether we would be able to infer these functions from data, we generate trajectories of length $T$ [See methods for details] and estimate $\mathcal{E}$ from the trajectories alone. We see an excellent agreement between the measured $\mathcal{E}$ and Eq. 74, while also maintaining agreement with $\hat{S}^{DBP}$ upon numerical integration of $\mathcal{E}$ (Supplementary Figure 3). This remains true for 3, and 4 dimensional simulations (Supplementary Figure 4), highlighting our ability to estimate $\mathcal{E}$ for high dimensional data.

We also investigate the effects of both the amount of data available and smoothing the covariance functions on $\hat{S}$ (Supplementary Figure 5). As expected, the spectral estimator becomes more accurate with more data, either by increasing $T$ or $N$. While S.Eq. 9 is exact for this model, noise in the estimated covariance functions leads to an overestimate of $\hat{S}$, which we can subtract. Smoothing systematically lowers $\hat{S}$, even after removing the systematic bias, by lowering the peaks in $\mathcal{E}$ [see Methods].

V. BRUSSELATOR DYNAMICS

The reversible Brusselator model we consider in this paper is defined by

\[
A \frac{k_1^+}{k_1} X; \quad B + X \frac{k_2^+}{k_2} Y + C; \quad 2X + Y \frac{k_3^+}{k_3} 3X,
\]

where $A, B, C$ are fixed external chemicals and the system is assumed to occur in a well-mixed vessel of volume $V$. Using mass action kinetics and writing lower-case letters as concentrations (e.g. $x \equiv X/V$), the macroscopic dynamics of the Brusselator are given by the coupled ODEs

\[
\dot{x} = k_1^+ a - k_1^- x - k_2^- bx + k_2^+ cy + k_3^- x^2 y - k_3^+ x^3
\]

\[
\dot{y} = k_3^- bx - k_2^- cy - k_3^+ x^2 y + k_2^+ x^3
\]

Detailed balance holds when each reaction rate in S.Eq. 77 is balanced, leading to the following equilibrium concentrations

\[
X_{eq} = A \frac{k_1^+}{k_1},
\]

\[
Y_{eq} = X_{eq} \frac{Bk_3^+}{Ck_2} = X_{eq} \frac{k_3}{k_3^+},
\]

where the first and second equation for $y_{eq}$ come from the $k_2$ and $k_3$ reactions, respectively. Using the two equations for $y_{eq}$ gives us the condition for detailed balance given in the main text, $Bk_3^+ k_1^- = Ck_2^- k_3^-$. The steady state values of $(x,y)$ are given by setting the deterministic equations to 0, giving

\[
x_{ss} = \frac{a k_1^+}{k_1^+}
\]

\[
y_{ss} = \frac{k_2^-bx_{ss} + k_3^-x_{ss}^3}{k_2^-c + k_3^-x_{ss}^2}
\]

The relaxation matrix, $R$, that defines the stability of the steady state is given by expanding the deterministic equations to first order around their steady state values

\[
R = \left( \begin{array}{cc}
\frac{\partial \dot{x}}{\partial x} & \frac{\partial \dot{x}}{\partial y} \\
\frac{\partial \dot{y}}{\partial x} & \frac{\partial \dot{y}}{\partial y}
\end{array} \right)_{ss} = \begin{pmatrix}
-(k_1^- + bk_2^+) + 2k_3^+ x_{ss} y_{ss} - 3k_3^- x_{ss}^2 & k_2^- c + k_3^- x_{ss}^2 \\
-bk_2^- - 2k_3^- x_{ss} y_{ss} + 3k_3^- x_{ss}^2 & -(k_2^- c + k_3^- x_{ss}^2)
\end{pmatrix}.
\]

The eigenvalues of $R$ are given by solving its characteristic equation, giving

\[
\lambda_{\pm} = \frac{\text{Tr}(R)}{2} \pm \left[ \left( \frac{\text{Tr}(R)}{2} \right)^2 - \text{det}(R) \right]^{1/2}
\]
VI. SIMULATION PARAMETERS

A. Gaussian Fields

Simulations of the Gaussian fields use an Euler Maruyama algorithm to integrate the equations of motion. Time and space are scaled by \( \tau = (Dr)^{-1} \) and \( \lambda = r^{-1/2} \). The simulation is performed on a periodic, 1 dimensional lattice.

| Name \( N_{sim} \) | Value | units | Description |
|------------------|-------|-------|-------------|
| \( dt \)         | 0.0001| \( \tau \) | simulation time step |
| \( t_{final} \)  | 50    | \( \tau \) | total simulation time |
| \( dx \)         | 0.1   | \( \lambda \) | spacing between lattice sites |
| \( N_{sites} \)  | 128   | 1     | number of lattice sites |
| \( \alpha \)     | \([0, 1, \ldots, 25]\) | \( \tau^{-1} \) | driving frequency |
| \( \sigma_\omega \) | 1.57  | \( \tau^{-1} \) | width of Gaussian used to smooth in the temporal dimension |
| \( \sigma_k \)   | 1.47  | \( \lambda^{-1} \) | width of Gaussian used to smooth in the spatial dimension |

B. Brusselator

Simulations of the Brusselator are done using a Gillespie algorithm \([5]\). Time is non-dimensionalized by \( \tau = 1/k_1^+ \).

| Name \( N_{sim} \) | Value | units | Description |
|------------------|-------|-------|-------------|
| \( k_1^+ \)      | 5000  | \( \tau \) | total time of simulation |
| \( k_1^- \)      | 0.5   | \( \tau^{-1} \) | forward reaction rate for reaction 1 |
| \( k_2^+ \)      | 2     | \( \tau^{-1} \) | reverse reaction rate for reaction 1 |
| \( k_2^- \)      | 0.5   | \( \tau^{-1} \) | forward reaction rate for reaction 2 |
| \( k_3^+ \)      | 2     | \( \tau^{-1} \) | reverse reaction rate for reaction 2 |
| \( k_3^- \)      | 0.5   | \( \tau^{-1} \) | forward reaction rate for reaction 3 |
| \( A \)          | 100   | 1     | number of chemical species in reaction volume |
| \( V \)          | 100   | 1     | reaction volume, used for calculated propensities in Gillespie algorithm |

The strength of external driving is given by \( \Delta \mu = (Bk_2^+ k_3^-)/(Ck_2^- k_3^+) \). Values of \( B \) and \( C \) are changed to give driving strengths \( \Delta \mu \in [-2, 8] \) with step size 0.1, while keeping the product \( \sqrt{(Bk_2^+ k_3^-)(Ck_2^- k_3^+)} = 1,000 \), where 1,000 is an arbitrarily chosen constant. The EPR plot in Fig. 3(c) uses a smoothing width of \( \sigma = 1.26 \), while the EPF plot shown in Fig. 3(b) uses a smoothing width of \( \sigma = 0.126 \) in order to better resolve the curve shape. Thus, the EPR measured by integrating the EPF in Fig. 3(b) is higher than the values shown in Fig. 3(c), although the shape of the curve remains unchanged.

C. Reaction-diffusion Brusselator

To add reactions to the Brusselator, we employ a compartment-based Gillespie algorithm. We non-dimensionalize time by \( k_1^+ \) and set the distance between each compartment to \( h = 1 \).
### Parameters

| Name  | Value | units | Description |
|-------|-------|-------|-------------|
| $N_{\text{sim}}$ | 10 | 1 | number of simulations per parameter set |
| $N_c$ | 64 | 1 | number of lattice sites |
| $D_X$ | $\lambda^2 \tau^{-1}$ | | diffusion constant of chemical species $X$ |
| $D_Y$ | $0.1 \lambda^2 \tau^{-1}$ | | diffusion constant of chemical species $Y$ |
| $t_{\text{final}}$ | 100 | $\tau$ | total time of simulation |
| $k_1^+$ | 1 | $\tau^{-1}$ | forward reaction rate for reaction 1 |
| $k_1^-$ | 0.5 | $\tau^{-1}$ | reverse reaction rate for reaction 1 |
| $k_2^+$ | 2 | $\tau^{-1}$ | forward reaction rate for reaction 2 |
| $k_2^-$ | 0.5 | $\tau^{-1}$ | reverse reaction rate for reaction 2 |
| $k_3^+$ | 2 | $\tau^{-1}$ | forward reaction rate for reaction 3 |
| $k_3^-$ | 0.5 | $\tau^{-1}$ | reverse reaction rate for reaction 3 |
| $A$ | 100 | 1 | number of chemical species in reaction volume |
| $C$ | 400 | 1 | number of chemical species in reaction volume |
| $V$ | 100 | 1 | reaction volume of each compartment, used for calculated propensities in Gillespie algorithm |

The strength of the driving $\Delta \mu$, and therefore the values of $B$ and $C$, is calculated in the same way as done for the Brusselator simulations without reactions. All subpanels involving the reaction-diffusion Brusselator use the same parameters with $\Delta \mu$ specified in the figure caption. Figures 4 and 5 uses a smoothing width of $(\sigma_\omega, \sigma_q) = (5, 0.5)$.

### D. Driven Brownian Particle

Simulations of the driven Brownian particle shown in the Supplementary Material use an Euler-Maruyama algorithm to integrate the equations of motion. Time is scaled by $\tau = \gamma/k$, which also scales the driving force. The variables have units of length and are scaled by $\lambda = \sqrt{D\gamma/k}$.

| Name  | Value | units | Description |
|-------|-------|-------|-------------|
| $N_{\text{sim}}$ | 64 | 1 | number of simulations per parameter set |
| $N_{\text{dim}}$ | 2 | 1 | number of dimensions of the simulation |
| $dt$ | 0.001 | $\tau$ | simulation time step |
| $t_{\text{final}}$ | 100 | $\tau$ | total simulation time |
| $\alpha$ | $[0, 1, \ldots, 10]$ | $\tau^{-1}$ | strength of driving |

Supplementary Figures 3 and 5 uses a smoothing width of $\sigma = 1.88$, while the smoothing width is varied in Supplementary Figure 4 as indicated by the colorbar.

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