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Inference of Onsager coefficient from microscopic simulations by machine learning ⊘

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ABSTRACT

Dynamic density functional theory (DDFT) is a fruitful approach for modeling polymer dynamics, benefiting from its multiscale and hybrid nature. However, the Onsager coefficient, the only free parameter in DDFT, is primarily derived empirically, limiting the accuracy and broad application of DDFT. Herein, we propose a machine learning-based, bottom-up workflow to directly extract the Onsager coefficient from molecular simulations, circumventing partly heuristic assumptions in traditional approaches. In this workflow, the Onsager coefficient is derived from the proposed DDFT-informed ordinary differential equation network, trained to replicate density evolution observed in Brownian dynamics (BD) simulations. We validate our method by studying the lamellar transition in symmetric diblock copolymer melts, where the DDFT model with the extracted Onsager coefficient precisely reproduces both the density evolution and interface narrowing predicted by BD simulations, thereby demonstrating the reliability of the present scheme. Meanwhile, our studies reveal the strong relevance of the Onsager coefficient with dynamic processes and identify the explicit connection between dynamic correlations, characterized by the correlation strength and correlation length, and the system parameters, e.g., the Flory–Huggins interaction parameter. We found that far from the transition point, the correlation that transmits the thermodynamic force into a density current is localized and strong, while close to the transition point, it becomes long-ranged but weak. Our approach aims to develop a more generalized framework to bridge more refined particle-based simulations to more coarse-grained field-based calculations, and the insights gained by using our approach could be extended to other non-equilibrium systems in polymer sciences.

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I. INTRODUCTION

Dynamic density functional theory (DDFT) describes the time evolution of the one-body density. It extends the classical density function theory^{1,2} by combining the free energy functional of the self-consistent field theory (SCFT)^{3,4} with a diffusive dynamical model for the polymer relaxation^{5,6} and gains reasonable success

in describing dynamic processes of polymer systems, for example, phase separations⁷⁻⁹ and order–disorder transitions.^{10–12} The general form of the DDFT evolution equation for an inhomogeneous multi-component polymer system is given by^{13–21}

$$\frac{\partial \phi_{\alpha}(\mathbf{r},t)}{\partial t} = \nabla \cdot \sum_{\beta} \int d\mathbf{r}' \Lambda_{\alpha\beta}(\mathbf{r},\mathbf{r}') \nabla \mu_{\beta}(\mathbf{r}',t), \qquad (1)$$

where $\phi_{\alpha}(\mathbf{r}, t)$ denotes the density field of component α at position \mathbf{r} and time t. $\mu_{\beta}(\mathbf{r}', t)$ is the chemical potential of component β at position \mathbf{r}' and time t, derived from the SCFT free energy functional $F\{\phi\}$ via $\mu_{\beta}(\mathbf{r}', t) = \delta F/\delta \phi_{\beta}(\mathbf{r}', t)$.^{3,4} $\Lambda_{\alpha\beta}(\mathbf{r}, \mathbf{r}')$ is a mobility matrix, also known as the Onsager coefficient, which relates the density current of the monomer α at position \mathbf{r} to the thermodynamic driving force $(-\nabla \mu_{\beta})$ on the monomer β at position \mathbf{r}' .^{14,22}

DDFT is characterized by its multiscale and hybrid nature in the sense that the free energy functional explicitly considers the microscopic topology of the polymer chains,^{3,4} while the mesoscopic dynamic equation has a heuristic origin as the Onsager coefficients $\Lambda_{\alpha\beta}$ are normally treated as free parameters.^{12,14,23–25} Different forms of the Onsager coefficient would lead to different transition pathways between two morphology phases. One typical example is the dynamics of the vesicle formation from spherical micelles, where the local Onsager coefficient predicted one single pathway during the self-assembly,²⁶ and in contrast, nonlocal Onsager coefficients suggested competing pathways,^{7,8} which were later confirmed by both experiments^{27–29} and simulations.^{30–33} Therefore, a proper choice of $\Lambda_{\alpha\beta}$ that accurately describes the nonlocal correlations in the system, is crucial for the applicability and validity of the DDFT.

Currently, the expressions of the Onsager coefficient are constructed in an empirical manner and with rational simplifications.^{12,14,23,34} In principle, the most rigorous way to derive the Onsager coefficient may start from the dynamic equations of the polymer monomer coordinates. Through a systematic coarsegraining scheme, the dynamics is projected on to the evolution of some collective variables, for example, polymer densities. Then, an explicit form of the Onsager coefficient could be extracted.³⁵ This is the basic idea of the Mori–Zwanzig formalism^{36,37} and the dynamic mean field theory.^{38,39} However, the finally obtained dynamic equation for the polymer density is guite complex due to the fact that contributions from different length and time scales are not effectively separated. This makes it difficult to obtain a definite and practically simple expression of the Onsager coefficient. To simplify the dynamic equation, Fraaije *et al.*^{23,40} considered the Rouse chain dynamics and obtained an evolution equation of the density being the same form as Eq. (1). In this formalism, the Onsager coefficient is proportional to the density correlation functions, which facilitates further simplification to the Onsager coefficient. There are a few choices of simplifications: one can neglect the density correlations to get a local dynamic model,²³ approximate the density correlation as the nonlocal Debye function,^{14,34} or obtain the external potential dynamics (EPD)¹⁴ by taking the translational invariant assumption. Further studies illustrate that the local DDFT typically overestimates the speed of phase transitions, whereas the nonlocal DDFT tends to underestimate it.^{9,12} To properly describe the multiscale behaviors, Qi and Schmid proposed a mixed scheme,¹² where the short scale movement of monomers represented by a local mobility function and large scale diffusion of polymer chains controlled by the Debye mobility function are coupled simultaneously. One main drawback of this mixed scheme is that a phenomenological parameter has to be introduced to tune the relative contribution from local and nonlocal dynamics, and this parameter is normally set empirically without solid physical considerations.²⁵ Due to its simplicity and extendibility, DDFT of Fraajie et al. and its variants have been widely used to study the dynamic behaviors of polymer systems."

Alternatively, one can obtain the Onsager coefficient by assuming Eq. (1) to be correct a priori and introducing further relations to determine $\Lambda_{\alpha\beta}$. Representative studies include the relaxation time approach $(RTA)^{25}$ and the dynamic random phase approximation (DRPA) theory,^{24,41} where the Onsager coefficients are all expressed in terms of the polymer chain structure factors. To get an analytical expression, the RTA and DRPA took the homogeneous state as the reference state and adopted free Gaussian chain approximations, which imply that they may fail when the system stays far away from the order-disorder transition point. It is interesting to note that in the RTA, a simulation method was also proposed; that is, from the simulation trajectory of polymer chains in a homogeneous reference state, the chain structure factor can be calculated directly, and then the Onsager coefficient is determined. Regarding molecular simulations to determine the Onsager coefficient, in fact, the Green-Kubo formalism gives a standard tool.42,43 However, numerical calculations and theoretical analysis demonstrated that the Green-Kubo relation is not suitable for evaluating q-dependent Onsager coefficient within the DDFT frame.²⁵

Although RTA and Green–Kubo formalism remain deficient in evaluating the Onsager coefficients,²⁵ molecular dynamic (MD) simulations have demonstrated their advantages, for example, they are able to establish directly a mapping between the microscopic information of the polymer systems and the Onsager coefficient, thus providing a feasible bottom-up scheme. However, it seems that few studies are progressed along this direction and lacking an efficient tool to deal with the large amounts of simulation data may be the main obstacle.

Fortunately, data-driven machine learning (ML) methods, as a general-purpose tool, have revolutionized many scientific and engineering fields.⁴⁴ It is an important research area to discover unknown governing parameters, such as transport and kinetic coefficients, in physical evolution equations from observational data, which typically falls under the category of inverse problems.⁴⁵ Currently, this type of research has received significant attention in various domains, including fluid dynamics,⁴⁶ heat transfer,⁴⁷ systems biology,⁴⁸ earthquake seismology,⁴⁹ and hydrology.⁵⁰ Even extracting governing equations has seen substantial progress.^{51,3} However, during the extraction of governing equations, no prior constraints are imposed to satisfy any physical principles, meaning the learned dynamics may lack clear physical structure and theoretical guarantees,53 thus posing a significant challenge to the physical interpretability of these equations. In the field of polymer science, especially phase-separated polymers, such research is still in its infancy. Ran et al.⁵⁴ have implemented ML techniques to infer the diffusivity in the constitutive equation from evolution trajectories of density. However, their data were obtained by solving analytical equations with given diffusion constants rather than through MD simulations or experiments. As a result, this approach may have trouble when confronted with inherent thermal fluctuations and noise in particle simulations or experimental systems.⁵⁵ Here, we propose a ML technique to extract Onsager coefficient functions from particle-based simulation data within the framework of DDFT, allowing field-based simulations to replicate the evolution observed in particle-based simulations. In addition, the extracted Onsager coefficients can facilitate the analysis of mesoscale correlations in the dynamic evolution of the system. This method offers a systematic framework for constructing mesoscale theory from microscopic simulations, avoiding most heuristic assumptions and unrealistic simplifications typically involved in theoretical derivations.

To exploit the microscopic information of molecular simulations, in this work, we combine the Brownian dynamics (BD) simulations with the DDFT [see Eq. (1)] constructing a hybrid scheme for the calculation of the Onsager coefficients. Concretely, we derive the Onsager coefficients from the DDFT-informed ordinary differential equation (ODE) network (DIO Net), which is trained to reproduce the density evolution from BD simulations within the DDFT framework. The obtained Onsager coefficients are assessed through the comparison of dynamic density functional calculations with the pure BD simulations, and the results show that they are physically reasonable and of high accuracy. In addition, our studies reveal the strong relevance of the Onsager coefficients with the dynamic processes, suggesting that the extracting of Onsager coefficients from the reference homogeneous states, which is normally done in practice, may have large deviations. By fitting the Onsager coefficient to a given form of correlation function, we find the explicit connection between the dynamic correlations and system parameters. Typically, we find that for a polymer system far from the transition point, the Onsager coefficient has a localized form with large magnitude, i.e., in this case, external forces inspire fast movement of nearby monomers, while close to the transition point, the Onsager coefficient becomes wide and weak; therefore, thermodynamic forces affect the motion of the whole chain in a long distance away but only weakly. The established formula of the Onsager coefficient facilitates the understanding of the microscopic origin of the nonlocal correlations in the system, and thus provides an opportunity to tune dynamic processes and other physical properties through the control of correlations. From the technical point of view, our scheme improves the performance of dynamic density functional modeling as it combines the advantages of the molecular simulations (fine-grained and accuracy) and the DDFT method (coarse-grained and efficiency). In fact, our study showed that to extract the Onsager coefficient, BD simulations with a short time interval are sufficient. Then, inserting the obtained Onsager coefficient into the DDFT allows a pure dynamic density field calculation, which is more efficient in comparison with BD simulations.

II. MODEL SYSTEM AND METHODS

To illustrate our combined scheme, let us consider the dynamics of morphology formation in diblock copolymer melts after a sudden quench from $\chi N = 0$ to a given $\chi N = 12-20$. Here, χN is Flory–Huggins interaction parameter and controls the incompatibility between different types of monomers.⁵⁶ In particular, the diblock copolymer melts are composed of symmetric flexible polymer chains, where the A block and B block have the same polymerization, i.e., $N_A = N_B$, with average monomer density ρ_0 and total polymerization $N = N_A + N_B = 40$ in a simulation box of size $L_x \times L_y \times L_z = R_g \times R_g \times 3R_g$, where R_g is the radius of gyration. Under this setup, the system would self-assemble from the initial homogeneous state into a lamellar structure with one period along the *z* direction.^{12,25} Please note that the box size is fixed for all different χN , which means that the system would be a bit frustrated at equilibrium and the lamellar spacing is not fully commensurable to the box length L_z .¹² Throughout this paper, lengths are represented in units of the radius of gyration R_g of an ideal chain, energies are in units of the thermal energy $k_B T$, and time is in units of $\tau = R_g^2/D_c$. Here, D_c represents the diffusion constant of one whole chain, which is derived from the monomer diffusivity D_0 via $D_c = D_0/N$.

In our study, we assume that the system is incompressible, i.e., at each location and any time, the volume fractions of A monomers and B monomers satisfy $\phi_A(\mathbf{r}, t) + \phi_B(\mathbf{r}, t) = 1$, which is commonly used in the previous studies.^{24,41,57-60} Then, the set of time evolution equations of the volume fractions ϕ_A and ϕ_B reduces to one,

 $\frac{\partial \phi(\mathbf{r},t)}{\partial t} = \nabla \cdot \int d\mathbf{r}' \Lambda(\mathbf{r},\mathbf{r}') \nabla \mu(\mathbf{r}',t),$

where

$$\phi(\mathbf{r}, t) \equiv \phi_{A}(\mathbf{r}, t) - \phi_{B}(\mathbf{r}, t),$$

$$\Lambda(\mathbf{r}, \mathbf{r}') \equiv \Lambda_{AA}(\mathbf{r}, \mathbf{r}') - \Lambda_{AB}(\mathbf{r}, \mathbf{r}'),$$

$$\mu(\mathbf{r}, t) \equiv \mu_{A}(\mathbf{r}, t) - \mu_{B}(\mathbf{r}, t).$$
(2)

In terms of the volume factions ϕ_A, ϕ_B and their corresponding conjugate potential fields ω_A, ω_B , the chemical potential difference μ can be expressed according to the SCFT as $\mu(\mathbf{r}, t) = -\chi N \phi(\mathbf{r}, t) + (\omega_B(\mathbf{r}, t) - \omega_A(\mathbf{r}, t))$. (See the Sec. S1 of the supplementary material).

Generally, the Onsager coefficient is nonlocal and historydependent;^{24,61} here as a proof-of-principle study and also to simplify the mathematical treatment, we have neglected the memory effect and also taken the translational invariant approximation to get $\Lambda = \Lambda(|\mathbf{r} - \mathbf{r}'|)$. The translational invariant approximation is valid when χN is close to 10.5 the order-disorder transition point.^{12,24,25,41,62} Now, we can conveniently formulate the DDFT equation [Eq. (2)] in Fourier representation³⁴ as

$$\frac{\partial \phi(\mathbf{q},t)}{\partial t} = -\mathbf{q}^2 \Lambda(\mathbf{q}) \mu(\mathbf{q},t), \qquad (3)$$

which serves as the basic equation in the current DDFT formalism. This implies from the theoretical aspect that single chain approximation and motion of center of mass are adopted, which fall into the framework of the theory by Fraaije *et al.*, i.e., memory effect is neglected.^{23,40} This should be valid in a later dynamic stage when the whole chain starts to migrate.

For molecular simulations, we adopt a Brownian dynamic scheme, where the interaction potentials are chosen as the Edwards type such that BD dynamics and the SCFT share the same form of Hamiltonian.⁶³⁻⁶⁵ This establishes the equivalence in principle between the particle-based BD simulation and the DDFT [Eq. (1)] in describing mesoscopic properties of the polymer systems and thus underlies our combination technique. In fact, the BD simulations deal with a compressible system, and to match the incompressible condition, the volume fractions obtained from the BD simulations are rescaled (see later part of the article for further information). The expression of the BD equation and other related details can be found in Sec. S3 of the supplementary material.

In principle, there is a mapping from microscopic dynamics (e.g., Newtonian dynamics and Brownian dynamics) to the dynamics of collective quantities (e.g. monomer density), which is normally referred to as the coarse-graining process. The structure of the obtained evolution equation of the collective quantities depends strongly on the form of the microscopic dynamics. However, a rigorous projection from BD dynamics for monomers to the dynamic density functional description would result in an evolution of polymer density with a complex form, which is hard to find a solution even numerically.^{38,39} In the present study, instead of performing the mapping, we adopt the assumptions by Fraaije *et al.* and construct an explicit and simple form of the density evolution equation with the memory effect being unfortunately lost.^{23,40} Therefore, the dynamic equation Eq. (1) gives a universal description of dynamics for polymers obeying Brownian dynamics given that the assumptions of Fraaije *et al.* hold.

On the basis of BD simulations and within the framework of DDFT, we propose a workflow shown in Fig. 1 to extract the Onsager coefficient. The main procedure goes as follows: initially, we run the particle-based BD simulations to generate the monomer trajectories for all monomers during the phase transition of the diblock copolymer melt from homogeneous to phase-separated lamellar states (middle-left of Fig. 1). The obtained monomer trajectories work as the original raw data.

A. Particle-to-field transformations

φ

Subsequently, to obtain a continuous and smooth density field (middle-center of Fig. 1), which is the collective variable that DDFT mainly concerns from discrete particle distributions, we employ the particle-to-field transformations. This is accomplished through two steps: (1) the particle-to-field convolution with a kernel (top left of Fig. 1) and (2) the average of the density field in a neighboring time interval (top right of Fig. 1).

In the particle-to-field transitions, the central quantity is the introduced smoothing function, which is the so called convolution kernel.^{51,66,67} The kernel serves as a weighting function that converts discrete particle locations in the system into continuous density fields, analogous to the point-spread function of a microscope. For example, with monomer positions denoted by $\mathbf{R}_{m,j}(t)$, an associated particle number density field $\phi(\mathbf{r}, t)$ can be defined by

$$(\mathbf{r},t) = \frac{1}{\tilde{\phi}(\mathbf{r},t)} \sum_{m=1}^{n_{\epsilon}} \sum_{j=1}^{N} I_{m,j} K(\mathbf{r} - \mathbf{R}_{m,j}(t)), \qquad (4)$$

where the normalization factor $\tilde{\phi}(\mathbf{r}, t) = \sum_{m=1}^{n_c} \sum_{j=1}^{N} K(\mathbf{r} - \mathbf{R}_{m,j}(t))$ represents the total density at position **r**, ensuring incompressibility,



FIG. 1. Sketch illustrating the main procedure of extracting the Onsager coefficient Λ in the framework of DDFT from particle trajectories generated by BD simulations. Initially, inputs are time-series data for particle positions $\mathbf{R}_i(t)$ obtained in BD simulations with the microscale resolution. Subsequently, density fields are obtained by employing convolution with a kernel [Eq. (4)] and density averaging [Eq. (7)]. Finally, the DDFT-Informed ODE Net is designed to determine the Onsager coefficient Λ from density fields. Herein, density $\phi(z,t)$ is defined as $\phi(z,t) =$ $\phi_{A}(z,t) - \phi_{B}(z,t)$ by using indicator function I_i , where $I_i = 1$ for monomer A, otherwise $I_i = -1$ for monomer B.

i.e., $\phi_A(\mathbf{r}, t) + \phi_B(\mathbf{r}, t) = 1$. The symmetric kernel $K(\mathbf{r})$ is centered at $\mathbf{r} = 0$ and is subjected to the normalization $\int d\mathbf{r} K(\mathbf{r}) = 1$, so that the total number of particles is recovered from $\int d\mathbf{r} \tilde{\phi}(\mathbf{r},t) = N n_c$. The indicator function $I_{m,j}$, which guarantees $\phi(\mathbf{r}, t) = \phi_A(\mathbf{r}, t)$ $-\phi_{\rm B}({\bf r},t)$, identifies the type of the *j*th monomer of *m*th chain. In particular, $I_{m,j} = 1$ for monomer A, otherwise $I_{m,j} = -1$ for monomer B. In general, the kernel function depends on the three dimensional coordinate r, which may be complex. However, the symmetry of the morphology phases (homogeneous state and lamellar phases) determines that the density varies effectively only along the z direction. Therefore, we define a one-dimensional density by taking the integration over x and y coordinates, i.e., $\phi(z,t)$ = $\iint dx dy \phi(\mathbf{r}, t)$. We should note that correlations still occur along the x and y directions, which are, however, not accessible because they give no contribution to the density evolution as the thermodynamic forces along these two directions are zero due to the homogeneity.

We have freedom to choose the form of the kernel function, which finally affects the smoothness of the density field. Here, we use a Gaussian kernel $K(z) \propto \exp\left[-z^2/(2\sigma^2)\right]$, where the kernel width σ is a free parameter (also see Sec. S4.1 of the supplementary material), and it determines the extent of coarse-graining. As shown in Fig. 2(a), when σ is small, the obtained density profile at equilibrium is rough with large fluctuations. This is because a narrow kernel could only map local particle distributions to the density. With the increase in σ , the kernel function becomes wider and more monomers are involved in the coarse-graining, so the density becomes smoother. It can be conceived that when σ is larger than the size of the system, all particles are taken into account, and the density approaches flat with the information of monomer distribution completely lost.

To find a compromise where the mapped density is sufficiently smooth, while information about monomer aggregations remains, we define a modified entropy $H(\sigma, t)^{68,69}$ to measure the quality of density conversion. $H(\sigma, t)$ has the meaning of information entropy (Shannon entropy),⁷⁰ and it encodes the spectral density,



FIG. 2. Influence of the Gaussian kernel width σ on the equilibrium density profiles ϕ_{eq} (a) and spectral entropy *H* (b) for the particle model data at equilibrium with $\chi N = 12$. Herein, H^{max} refers to the value of *H* at $\sigma = 0.0003R_g$ used to normalize *H* and show the proportion of information retained for different values of σ compared to $\sigma \rightarrow 0$. The dashed red line in panel (b) indicates the spectral entropy corresponding to the equilibrium density profile at $\chi N = 12$ as derived from self-consistent field theory.

i.e., distribution of power in the frequency domain, into one value, i.e.,

$$H(\sigma, t) \sim -\sum_{\mathbf{q}} S(\sigma, t, \mathbf{q}) \log_2 S(\sigma, t, \mathbf{q}),$$
(5)

where $S(\sigma, t, \mathbf{q})$ is the amplitude of the density fields $\phi(\sigma, t, z)$ in Fourier space,

$$S(\sigma, t, \mathbf{q}) = \left| \int dz \phi(\sigma, t, z) \exp\left(2\pi i \mathbf{q} \cdot z\right) \right|^2.$$
(6)

This type of Shannon entropy is widely used across various fields, such as brain-computer interface^{68,71} and machinery condition monitoring,^{69,72} to understand the changes in sequence data. According to the definition, the modified spectral entropy can yield a larger value when the amplitude distribution is flat or the density peak is very high; otherwise, it gets a smaller one.

Figure 2(b) shows the scaled spectral entropy H/H^{max} of the equilibrium density $\phi_{\rm eq}$ with respect to the extent of the coarsegraining measured by σ . As expected, with the increase in σ , the density profile becomes smoother since more microscopic information about the polymer states is lost, and the spectral entropy exhibits a monotonic decrease. The sharp decrease in H in the small σ region corresponds to the process of smearing of particle locations, while that in the large σ region is related to the flattening of the density field. In contrast, in the intermediate region, where σ spans roughly from $0.01R_g$ to $0.1R_g$, H decreases quite slowly approaching a constant. This indicates that, in the plateau region, the density profile is insensitive to the coarse-grain extent σ . Therefore, any σ situated within the plateau region gives us an appropriate choice to generate a density profile, which is very smooth and simultaneously contains sufficient information to accurately capture the density evolution. In fact, we have compared the densities produced by several chosen σ within the plateau H region, and almost exactly the same density profiles are observed showing no influence of the coarse-graining extent (see Fig. S5 in the supplementary material for details). Without loss of generality, we set $\sigma = 0.05R_g$ in the dynamic evolution process for deriving a continuum model from the simulation data in the following calculations.

To further refine the smoothness of the density field, after the particle-to-field conversion, we also perform a time average of the density from different density trajectories in a short neighboring time interval. Mathematically, the averaging can be formulated as

$$\phi(z,t_i) = \frac{1}{n_T} \sum_{t_j \in U_{t_i}} \phi(z,t_j), \qquad (7)$$

where U_{t_i} denotes a time range $[t_i - \varepsilon, t_i + \varepsilon]$ corresponding to about 1000 density samples ensuring minimal density variation and sufficient data for averaging, and n_T is the number of density samples in U_{t_i} . The density fields obtained in this way are much smoother, as confirmed by the comparison of density profiles before and after such averaging [see Figs. 3(a) and 3(b)]. This average is commonly carried out in the calculation of correlation functions to reduce the statistical errors and is referred to as the pre-average or block average.⁷³ Here, such average resembles the ensemble average and in addition, avoids unnecessary computational burdens. In fact, in



FIG. 3. Density profiles after the smoothing with the Gaussian kernel (a) and further by the average of density trajectories sampled within a short time interval (b) at different time. The Flory–Huggins parameter is chosen as $\chi N = 12$.

most cases, this average is necessary, especially in a dilute system, because the particle-to-field mapping cannot get a perfectly smooth density upon just one instant chain configuration. Even small fluctuations in the density profile would be strongly enlarged when evaluating the chemical potential through the density inversion in getting the auxiliary potential, as the profile of auxiliary potential is susceptible to the density distributions, which may result in a numerical failure in the subsequent calculations. For the present system, our calculations show that this pre-average is necessary, otherwise the large fluctuations in $\phi(z, t)$ would not lead to a meaningful Onsager coefficient.

B. Inference of the Onsager coefficient by machine learning

Finally, the density fields generated in this way are used for model training to obtain the Onsager coefficient Λ by using DDFT-Informed ODE Net (bottom of Fig. 1), inspired by DDFT. More details are given in the following. Once the density fields are obtained, while, in principle, it is possible to directly solve the Onsager coefficient Λ by Eq. (3), this is not a trivial task in practice. The smoothness of density $\phi(\mathbf{r}, t)$ at a given time tdoes not consequently result in a continuous evolution of $\phi(\mathbf{r}, t)$, i.e., non-smooth $\partial_t \phi(\mathbf{r}, t)$. In addition, the chemical potential μ , derived from SCFT, is sensitive to density fields. The mathematical features of these quantities related to the continuity and noise severely hinder the computation of the Onsager coefficient Λ against overfitting.

Therefore, as shown in the bottom panel of Fig. 1, we specifically design a DDF-Informed ODE Net (DIO Net), which consists of two sub-networks, the ODE Net and the fully connected Neural Net. ODE Net, first proposed by Chen *et al.*,⁷⁴ is a deep learning algorithm that blends neural networks, which learn the time derivatives, with time integration methods, such as the Runge–Kutta method,⁷⁵ and facilitates the learning of time-continuous dynamics, represented as a system of differential equations, solely from discrete-time observations. Herein, we use it to obtain the time partial derivative of the density $\partial_t \phi(\mathbf{r}, t)$ from discrete-time density fields. Meanwhile, the fully connected Neural Net is used to describe the Onsager coefficient Λ by minimizing the DDF loss and ODE loss simultaneously. In particular, the ODE Net is pre-trained, i.e., initially training the ODE Net solely, to facilitate faster convergence. Once well trained, $\Lambda^{\text{ML}}(\mathbf{r}, \mathbf{r}')$ obtained from this ML model can be incorporated into the DDFT equation, enabling the density to evolve from the initial time. More details about the model structure and training process are provided in the Secs. S5.1 and S5.2 of the supplementary material.

In most cases, physics-informed neural network (PINN), a type of universal function approximator embedding the knowledge of physical laws described by partial differential equations into the learning process, is used to compute the derivatives of functions.^{54,76,77} However, in our study, we utilize ODE Net instead of PINN. This choice is primarily because ODE Net models time series based on differential equations, whereas PINNs merely fit the time series, often leading to overfitting of data. Moreover, ODE Net is capable of handling non-regularly spaced intervals and incomplete measurement data.⁷⁸ This capability facilitates the extension of the model to experimental measurements, thereby addressing potential issues of missing data.^{78–80}

We extract the time-dependent density profile in a time window of $[t_0, t_e]$ from BD simulations for the model training. The time duration $\Delta t = t_e - t_0$ is about one-quarter of the region where the density profile changes significantly. Here, t_0 denotes the initial time of the sampling interval, which is roughly the time when the interface begins to form and will be discussed in detail in the following. Within this time window, due to the time step $dt_{BD} = 0.001/N$ of BD simulations, there are about ten thousand time points available. However, in practice, such a large dataset is unnecessary since the variation of density fields between consecutive time points is minimal. Therefore, we select approximately $N_t \approx 400$ evenly spaced time points within this time window to train the model. Notably, we found that the final results are not highly dependent on the specific initial time t_0 or the time duration Δt ; any sub-time period between the initial time t_0 and the time when the density fields just reach at the equilibrium state can also produce satisfactory results, which will be discussed in detail later.



FIG. 4. Time evolution of the normalized spectral entropy H_{norm} , which describes the rate of evolution and the maximum density ϕ^{max} . Herein, $H_{\text{norm}}^{\text{max}}$ refers to the value of H_{norm} at t = 0, used to normalize H_{norm} and show the proportion of information retained for different values of t compared to t = 0. The dashed red line indicates the initial time $t_0 = \tau$. The parameter $\chi N = 12$ is chosen.

In the early stages of the structure evolution, due to the subdiffusive segmental dynamics, the Onsager coefficient is timedependent and results in significantly faster structure forma-tion dynamics in $t \ll \tau$ than in $t > \tau$.²⁴ To extrapolate a timeindependent Onsager coefficient with ML, we determine the initial time t_0 with the normalized spectral entropy H_{norm} . The spectral entropy H_{norm} is based on Eq. (5) and introduces a normalization factor $\left[\int d\mathbf{q}S(t,\mathbf{q})\right]^{-1}$ for *S* to eliminate the influence of the absolute value of density on entropy, highlighting the changes in the shape of the density profiles.^{68,69} As shown in Fig. 4, at the initial stage, the shape of density changes rapidly and thus results in a rapid decrease in H_{norm} . Later, a plateau of H_{norm} emerges, indicating that the shape of the density becomes nearly constant. Therefore, for our system, we roughly choose the turning point of H_{norm} as the initial time for learning the Onsager coefficient, i.e., $t_0 = \tau$ at $\chi N = 12$. For a deep quench, the density changes fast, and $H_{\text{norm}}/H_{\text{norm}}^{\text{max}}$ saturates to the final stable value in an early time, so in practice, we set a smaller t_0 with the increase in χN .

III. RESULTS AND DISCUSSION

For the assessment of our scheme, we first compare the result of Onsager coefficient Λ^{ML} extracted using the present method and that from several other models. Figure 5(a) presents the Onsager coefficients as a function in Fourier space, and $q = 2\pi k/L_z$, where k is an integer. For the Debye model,¹⁴ we take $\Lambda^{\text{Debye}} = g_{\text{AA}} - g_{\text{AB}}$, and $g_{\alpha\alpha}(q) = g_{\text{D}}(f,x)$, $g_{\text{AB}} = \frac{1}{2}[g_{\text{D}}(1,x) - g_{\text{D}}(f,x) - g_{\text{D}}(1 - f,x)]$, where $x = q^2 R_g^2$, $g_{\text{D}}(f,x) = \frac{2}{x^2}(fx + e^{-fx} - 1)$ is the Debye function,³⁴ with f being the fraction of block α . The mixed scheme is a variant of that proposed by Qi and Schmid,¹² where incompressible conditions are imposed to get an analytical equation, and the corresponding Onsager coefficient in Fourier space is written as

$$\Lambda^{\text{mixed}}(q) = G_{\text{AA}}(q) - G_{\text{AB}}(q) + \frac{1}{2} - \frac{1}{2}\Xi(q).$$
(8)

Here, $\Xi(q)$ is the predefined filter function expressed in real space as $\Xi(r) = (2\pi\xi^2)^{-3/2} \exp(-r^2/2\xi^2)$, which is used to separate the

thermodynamic forces into a "coarse-grained" nonlocal part and a "fine-grained" local part, and ξ is a tunable parameter controlling the length scale to crossover between local $(\Lambda^{\text{Local}}(|\mathbf{r} - \mathbf{r}'|))$ = $0.5\delta(|\mathbf{r} - \mathbf{r}'|)$ and nonlocal Debye dynamics. In terms of the filter function, $G_{\alpha\beta}$ reads $G_{\alpha\beta}(|\mathbf{r} - \mathbf{r}'|) = \int d\mathbf{r}'' g_{\alpha\beta}(|\mathbf{r} - \mathbf{r}''|) \Xi(|\mathbf{r}'' - \mathbf{r}'|)$. It is obvious that as ξ approaches zero, the filter function $\Xi(r)$ becomes the delta function, meaning that on all length scales the dynamics are coarse grained, and the mixed model recovers the Debye description. On the other limit, when ξ is very large, Ξ becomes wide and small, and $\Lambda^{\text{mixed}}(q) \rightarrow 0.5$, and then local dynamics dominants (see Fig. S1 in the supplementary material). This shows the strong ability for the mixed model to describe the local movement of monomers and diffusion of the whole chain given that ξ is properly set, which is normally difficult as it depends on particular polymer systems. The data of the Onsager coefficient from the RTA, i.e., Λ^{RTA} , are taken from the work of Mantha et al.²⁵ under the same parameters (see Sec. S2.1 of the supplementary material for more detail). It has a finite value at q = 0 indicating the existence of finite time for long-wave compositional fluctuations. In contrast, the mixed and Debye models predict the divergence of relaxation time.25

The Onsager coefficient obtained from machine learning is close to that of the Debye model at large q and increases faster when q becomes smaller, leading to $\Lambda^{ML}(q=2) > \Lambda^{Debye}(q=2)$. However, the value of Λ^{ML} at q=0 cannot be obtained by the present machine learning scheme because the data fitting fails to work at q=0 as the dynamic equation [Eq. (3)] gives no restriction on $\Lambda^{ML}(q=0)$. The tendency of the profile of Λ^{ML} approaching q=0 can be predicted by taking a smaller $\Delta q = \frac{2\pi}{L_z}$, i.e., considering a system with a larger size L_z , which is not studied here.

We also plot in Fig. 5(b) the Onsager coefficients in real space, transformed by using fast Fourier transforms from those in Fourier space, in order to have a direct impression of the form of nonlocal correlations. Still, the profile of Λ^{ML} is quite close to that of Λ^{Debye} , except that Λ^{ML} has the freedom of a vertical shift as $\Lambda^{ML}(q=0)$ has not been determined. The Λ^{RTA} decreases non-monotonically to zero with increasing the distance, which is expected, since Λ^{RTA} calculated through all monomer positions characterizes the structure of the copolymer chains in the homopolymer state. Differing



FIG. 5. Onsager coefficient obtained by using different methods as denoted in Fourier space (a), real space (b), and the evolution of maximum density difference during the phase transition process predicted by BD simulations and DDFT with different forms of the Onsager coefficient (c). The cyan line in panel (c) is the result from the BD simulations. Herein, the incompatibility parameter is set to $\chi N = 12$, and in the mixed scheme, we choose $\xi = 0.5$ and $\Lambda^{\text{Local}} = 0.5\delta(|z - z'|)$. Λ is defined as $\Lambda = \Lambda_{\text{AA}} - \Lambda_{\text{AB}}$.

from Λ^{RTA} , Λ^{ML} decreases monotonically, although Λ^{ML} also explicitly considers all monomer positions. The difference between Λ^{ML} and Λ^{RTA} may imply that the Onsager coefficient strongly depends on the state of the system in the transition processes, a homogeneous treatment of the system could result in large deviations in the Onsager coefficients from true ones.

By inserting these Onsager coefficients back into the dynamic density functional equation, we can get the density evolution. To quantify the phase transition process from a homogeneous state to the lamellar state, we focus on the evolution of the density maximum, which is defined as $\phi^{\text{max}} \equiv \phi_A^{\text{max}} - \phi_B^{\text{min}}$. Figure 5(c) shows ϕ^{max} as a function of times, and for comparison, the result from pure BD simulations is also plotted. Please note that as the machine learning only considers some parts of the evolution trajectories later than $t > \tau$, we discard density evolution earlier than $t = \tau$. With the passage of time, all models predict that ϕ^{max} saturates to the same equilibrium value, but they have different rates of ordering. The local dynamics, which emphasizes local arrangements, overestimates the phase ordering speed, while the Debye dynamics, which mainly represents the large-chain behavior, underestimates the ordering rate.^{9,12} For the mixed model at $\xi = 0.5$, the ϕ^{max} grows still faster compared to the BD simulation result, indicating improper interpolating between the local and Debye dynamics. A nice match between the mixed model and the BD simulations can be expected when ξ decreases to a sufficiently small value. Compared to the machine learning method, the RTA overestimates the phase transition rate, which again demonstrates that to obtain a more exact Onsager coefficient, one has to consider the polymer conformations during the phase transitions.

To further investigate the influence of the transition trajectories on the Onsager coefficient, we vary the Flory–Huggins parameter χN and calculate the corresponding Λ^{ML} in each case. Depending on χN , the final equilibrium lamellar phases are different, for example, they have different interfacial widths, although initiated at the same homogeneous state. This means that for different χN , chain conformations and bond ordering extent are different when the systems have evolved the same time duration representing diverse transition trajectories. Our scheme effectively takes into account the transition trajectories by sampling the system for machine learning in a short time interval during the transitions rather than only considers the chain conformations at the initial homogeneous states.

Figure 6 shows the profiles of the Onsager coefficient in Fourier and real space with varying χN . Differences are obviously seen in the profiles of Λ for different χN . In general, with the increase in χN , Λ^{ML} becomes larger almost for each mode q, which suggests that the mobility of the polymer chains at different length scales is all enhanced under deeper quench. This matches the general idea of critical slowing down, as inversely when χN decreases to approaching the critical point (the exact location of the critical point is not identified), the chain mobility becomes weaker and tending flatter [see Fig. 6(a)]. In real space, as shown in Fig. 6(b), $\Lambda(z)$ becomes thin and sharp as χN increases, which illustrates strong correlations at short lengths. This is consistent to the fact that the interfacial length is small for a large χN (see Fig. 7). It should be noted that due to undetermined vertical shift, the absolution value of $\Lambda(z)$ may not be trustable. Still, the strong dependence of Λ over χN confirms



FIG. 6. Onsager coefficient Λ obtained by the present machine learning scheme represented in Fourier space (a) and real space (b) for several chosen χN as denoted.



FIG. 7. Interfacial width *W* as a function of time in double-logarithmic representation, as obtained by the BD simulation (solid lines) and DDFT models (dashed lines) with the Onsager coefficient given by the machine learning scheme for different χN . In particular, the values of χN are 12, 12.4, 13, 14, 15, 17, and 20, respectively. The power law $W \propto t^{-2}$ is shown as a guide for the eye in the plots.

the necessity of involving transition trajectories when calculating the Onsager coefficient.

As a simple application, we use the present scheme to study the process of interfacial narrowing during phase transitions. After the quench, polymer chains gradually move to form monomer A rich and B rich regions from randomly distributed state at the start, and the density profile changes from flat to curved with a sharp change of the density value at the interfacial regions. To quantify the interfacial region, we define the interfacial width W as the inverse of the maximum slope of the density profiles ϕ_A , which is located at the center of the interface regions (other definitions are possible, but the results qualitatively hold).¹² Figure 7 shows the change of interfacial width W as a function of time for different χN parameters obtained by both BD simulations and DDFT method. In the DDFT scheme, the Onsager coefficients were extracted by the fitting the BD data within a short time interval. The starting point t_0 and the duration Δt of the time interval depend on the parameter χN ; roughly speaking, it spans about 1/4 of the region where the interfacial width



FIG. 8. (a) Numerical fitting of the learned Onsager coefficient in real space. The data (solid lines) are fitted to a stretched exponential function (dashed lines), $\Lambda(\Delta z) = a_0 \exp\left(-\left(\frac{\Delta z}{r_0}\right)^{\beta_0}\right)$, with length scale r_0 and stretching parameter β_0 . (b) Effective correlation length r_{eff} and the interfacial width *W* as a function of the incompatibility parameter χN .

changes dramatically. For example, in the case of $\chi N = 12$, we set $t_0 = 1\tau$, and the time interval ranges from $t = 1\tau$ to $t = 2\tau$, and for $\chi N = 20$, $t_0 = 0.175\tau$ and the time duration for sampling $\Delta t = 0.1\tau$. The curves show that except for the early times and the saturating stage, the decrease in *W* for different χN follows an apparent scaling relation $W(t) \approx t^a$ at the intermediate times with the exponent $\alpha \approx 2.0$. Although $\Lambda^{\rm ML}$ in the DDFT is obtained from limited amount of data sampled in early times, they correctly predict the evolution of systems afterward. This can be confirmed from Fig. 7 that the evolutions of *W* with respect to time from the pure BD simulation and the DDFT scheme almost overlap each other for each given χN . We will come back to discuss the influence of t_0 and time duration later.

From the Onsager coefficient, we can learn further information about the correlation in the system, and we call such correlation the dynamic correlation as it specifies a region within which a thermodynamic force can induce a flow. Normally, with respect to the distance, the strength of correlation exhibits a decay, so to model such decay, we introduce a "stretched" exponential function, which has the form of $\Lambda^{\text{fit}}(z) = a_0 e^{-(z/r_0)^{\beta_0}}$. Here, a_0 is a prefactor, r_0 represents a characteristic length scale, and β_0 describes the deviation of the correlation function from exponential decay. We obtain these parameters by fitting the data Λ^{ML} to Λ^{fit} in real space. From the correlation curve, we calculate an effective correlation length $r_{\text{eff}} = a_0^{-1} \int_0^{\infty} dz \Lambda_{\text{fit}}(z) = \frac{r_0}{\beta_0} \Gamma(1/\beta_0)$,⁸¹ Γ being the gamma function. From Fig. 8(a), we see that in a wide range of χN , Λ^{ML} fits reasonably well to Λ^{fit} .

Figure 8(b) shows the effective correlation length $r_{\rm eff}$ as a function of the incompatible parameter χN , and for comparison, the data of interfacial length W are also presented. It can be seen that the effective correlation length is comparable with the interfacial width in order of magnitude, although the concrete value of W depends on its definition. In addition, $r_{\rm eff}$ is smaller than the mean radius of gyration of the polymer $R_{\rm g}$. However, with the decrease in χN , $r_{\rm eff}$ grows fast, and it is conceivable that $r_{\rm eff}$ becomes larger than $R_{\rm g}$, when the system is close enough to the critical point. Similar to the case of W, with the increase in χN corresponding to a deeper quench, the correlation length becomes smaller. This indicates that far from



ARTICLE

FIG. 9. (a) Onsager coefficient Λ^{ML} expressed in Fourier space for different training time windows. The inset gives the corresponding plots of Λ^{ML} in real space. (b) The time evolution of the maximum density ϕ^{max} depicted according to BD simulations (cyan line) and DDFT calculations with various training time windows as indicated. The color coding in panel (b) also refers to panel (a).

the transition point, the influence of external forces becomes localized but strong, i.e., only chains nearby move fast. On the contrary, approaching the transition point, the thermodynamic force could inspire the movement of chains in a long distance away, but the response is weak, so they move slowly. We expect that such features of the dynamic correlations may have consequences on the transition paths under different conditions, which require further investigations.

For all χN that we have considered (i.e., $12 \le \chi N \le 20$), the stretching parameter β_0 is found to range between 1 and 2 with no monotonic dependence on χN (see Table S1 of the supplementary material for the values of β_0). The parameter β_0 larger than one means that the dynamic correlation decays faster than the single exponential form, which is also consistent to the fact that $r_{\rm eff} < r_0$. The short-ranged type of correlations between thermodynamic forces and flows may originate from the BD description of the polymer chains, where long-ranged hydrodynamic interactions are not involved.

As we have mentioned before, to extract the Onsager coefficient, we sampled the density obtained by BD simulations within a short time interval between t_0 and t_e , where t_0 and t_e depend on χN , and $\Delta t = t_e - t_0$ approximately covers 1/4 of the sharp phase transition region. Now, we discuss the effect of t_0 and t_e on Λ^{ML} and the density evolution. As an example, we consider the case of χN = 12, and in previous calculations, we have chosen $t_0 = 1\tau$ and $t_{\rm e} = 2\tau$. For comparison, we choose another two sets of parameters, and for the first set $t_0 = 1\tau$ and $t_e = 4\tau$ meaning a longer sampling duration, while the other set $t_0 = 2\tau$ and $t_e = 3\tau$ corresponding to a different starting time. We plot the profiles of Λ^{ML} as a function of q and z (the inset) for these different training time windows shown in Fig. 9(a). These curves in Fourier and real space nearly completely coincide with just minor deviations. It should be noted that $\Lambda^{\overline{ML}}$ definitely has a close relation with the sampling window, for example, Λ^{ML} would have a different form if one samples the densities in a time interval with t_0 and its width approaching zero, i.e., near the initial homogeneous state. However, our study demonstrates at least that there exists a short time window within which the evolution of densities can generate an Onsager coefficient of high accuracy and stability.

30 April 2025 09:02:5

Next, we solve the dynamic density evolution equation by inserting these Onsager coefficients into the dynamic equations and obtain the growing of ϕ^{max} from the initial homogeneous state to the final lamellar phase. Figure 9(b) shows the profiles of ϕ^{max} with respect to time for pure BD simulations and DDFT. The curves corresponding to different sampling intervals almost get overlapped, and they also reproduce well the results from BD simulations. Thus, besides the fact that these Onsager coefficients do not depend on the initial time and the size of the sampling time intervals, they can also correctly describe the dynamic propagation of polymer chains, which confirms the robustness of our machine learning scheme.

In addition, Fig. 9 shows that our machine learning scheme could be more efficient than the pure BD simulations while still at the same level of accuracy. In general, particle-based BD simulations are time consuming, as the polymer systems, especially polymer melts, contain quite a large number of monomers, and dealing with all these monomers takes a lot of time. On the other hand, the DDFT runs fast, as the time cost there is solely relevant to the total number of cells that the system partitions to, which is normally not that large, for instance, in a one-dimensional system. If we construct the Onsager coefficient from BD simulations but within a short time interval, and then perform DDFT calculations instead, we can reach a high performance modeling with high accuracy and efficiency. The speedup could be one or two orders of magnitude (see Sec. S5.5 of the supplementary material for further information).

IV. SUMMARY AND REMARKS

To summarize, in the present work, we have proposed a bottom-up scheme to give an explicit expression for the Onsager coefficient within the DDFT framework. Our scheme relies on the mapping from the particle-based trajectories (obtained by separate BD simulations) to the propagation of density fields, which is accomplished through a DDFT-informed machine learning neural network. To exemplify the application of our scheme, we focus on the phase transition of a symmetric diblock copolymer system from the initial homogeneous state to the final lamellar phase after a quench, and have mainly the following conclusions:

- The dynamic density functional equation with the Onsager coefficient extracted from the BD data can reproduce reasonably well the evolution of polymer densities as well as the narrowing of the phase interfaces predicted by the pure BD simulations during the phase transition process demonstrating the accuracy of the obtained Onsager coefficient and the reliability of the conducted machine learning procedure.
- As the particle moving trajectories with a time intervals are considered, our scheme captures at least partially the process-dependent nature of the Onsager coefficient. This matches the result that the expression of the Onsager coefficient depends strongly on the quench depth χN , which controls the transition path.
- In addition to the diffusivity, which is represented by the magnitude of the Onsager coefficient, the range where a thermodynamic force inducing a diffusive flux, which is

quantified by the correlation length, can be obtained by fitting the Onsager coefficient to a given form of correlation function. By doing so, we reveal the dynamic coupling properties of the polymer systems under different conditions. For example, when the system is far from the transition point, the influence of external forces becomes localized but strong, while close to the transition point, the association of the thermodynamic force covers a large range but weak. We expect that the analysis of these dynamic correlation functions would help specify the transition path ways.

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Although the present studies focused on the order-disorder transition of a simple diblock copolymer melt, it is straightforward to extend our scheme to study the dynamic processes of more complex systems. The obtained explicit form of the Onsager coefficient would help understand the connection between the microscopic structure of the polymer chains, external controlling conditions, and the real transition path ways between different phases. In addition, to decrease the number of free variables and to ease the machine learning treatment, we have imposed the incompressible condition, which is in fact not a principle requirement by the scheme. The current BD simulations deal with compressible systems with the compressibility characterized by the Helfand parameter κ , and we have set $\kappa N = 10$ in the simulations reflecting moderately compressible situations. The incompressible condition corresponds to the limiting case in the BD simulations when κ increases to infinity.

The Onsager coefficient extracted by the present scheme does not explicitly depend on time. However, in practice, phase transitions represent a nonequilibrium process, and the evolution of morphologies may strongly depend on the history of the self-assembly. This means that the Onsager coefficient should be a function of both positions and time. In the present study, the Onsager coefficient shows no clear associations with the time intervals chosen for data sampling, which may indicate that the memory effects are not that crucial here or they are averaged out in some way.²⁵ We believe that memory effects are important in determining dynamic processes and the simulation study here provides a basic step for the further improvement of machine learning schemes to involve the time correlations in the Onsager coefficient.

Our approach establishes a direct linkage between particle simulations and field-based models. Field-based models primarily focus on the dynamics of collective variables within the system (e.g., density), thereby offering a more direct and effective description of the system's long-term structural evolution at the mesoscopic scale. These models are highly dependent on the characterization of the system's microscopic details, such as the topological structures of chains and transport coefficients of the monomers. However, these coefficients are typically derived through theoretical methods under specific physical conditions, where the approximations employed during the derivation are often excessively simplified. As a result, the Onsager coefficients derived from theoretical methods may not accurately reflect the microscopic transport behaviors of systems, leading to significant discrepancies in the dynamic processes and structural evolution pathways, or even errors. To address this issue, our research presents an effective computational framework that directly derives the relevant dynamic coefficients from particle simulation trajectories. This not only mitigates the flaws

ARTICLE

inherent in traditional theoretical derivations but also provides new insights into dynamics. Similarly, this methodology can be applied to connect other scales, paving the way for the future use of multiscale simulations to study more complex nonequilibrium dynamical problems.

SUPPLEMENTARY MATERIAL

The supplementary material is available free of charge and includes the methods of SCFT, DDFT, BD simulation, and machine learning.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Kaihua Zhang: Formal analysis (equal); Investigation (equal); Methodology (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). Shuanhu Qi: Conceptualization (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Supervision (equal); Writing – original draft (equal); Writing – review & editing (equal). Yongzhi Ren: Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Writing – review & editing (equal). Jiajia Zhou: Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Writing – review & editing (equal). Jiajia Zhou: Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Writing – review & editing (equal). Supervision (equal); Investigation (equal); Methodology (equal); Supervision (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Supplementary Material for Inference of Onsager Coefficient from Microscopic Simulations by Machine Learning

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Contents

S1	Self-Consistent Field Theory	3
S2	Dynamic Density Functional Theory	5
	S2.1 Schemes for Onsager coefficient	5
	S2.2 Settings of DDFT calculation	9
	S2.3 Influence of the absence of wavenumber $ {\bf q} <2$ in $\Lambda({\bf q},t)$ on $\Lambda(z-z' ,t)~$.	9
S3	Brownian Dynamics Simulations	11
	S3.1 Simulation method	11
	S3.2 Influence of compressibility parameter on density	12
$\mathbf{S4}$	Coarse-grained Density Fields	14
	S4.1 Kernel coarse-graining with periodic boundaries	14
	S4.2 Independence of Gaussian kernel width σ on the density fields $\ldots \ldots \ldots$	14
$\mathbf{S5}$	Machine Learning Methods	16
	S5.1 Architecture of the DDFT-Informed ODE Net	16
	S5.2 Training the DDFT-Informed ODE Net	17
	S5.3 Making predictions using Λ^{ML}	18
	S5.4 Additional results for numerical fitting	19
	S5.5 Speedup factor of DDFT simulations	19

S1 Self-Consistent Field Theory

In this study, we employ the self-consistent field theory (SCFT), which is one of the most powerful equilibrium theories for inhomogeneous polymer systems,^{1,2} to construct the free energy functional in DDFT equations. In this section, we briefly summarize the main equations adjusted to our system.

The system with a volume V contains n monodisperse chains of symmetric AB diblock copolymers with an average monomer density ρ_0 and total polymerization $N = N_{\rm A} + N_{\rm B}$. The block copolymers are modeled as continuous Gaussian chains³ with the contour length parameterized by the continuous variable s, which ranges from 0 to 1, and the interactions between the A and B components are modeled by the combined Flory-Huggins parameter χN . Therefore, the dimensionless free energy functional $F\{\phi_{\alpha}(\mathbf{r})\}$ in units of thermal energy $k_{\rm B}T$ can be expressed as.²

$$F = \frac{\rho_0}{N} \left\{ \int d\mathbf{r} [\chi N \phi_{\rm A}(\mathbf{r}) \phi_{\rm B}(\mathbf{r})] - \sum_{\alpha = {\rm A}, {\rm B}} \int d\mathbf{r} \phi_{\alpha}(r) \omega_{\alpha}(\mathbf{r}) - V \ln Q \right\}$$
(S1)

where ϕ_{α} and ω_{α} represent the normalized density field and conjugate field of monomer α , respectively, and Q is the single chain partition function.

The conjugate fields ω_{α} can be calculated through the implicit relationship

$$\phi_{\alpha}(\mathbf{r}) = \frac{N_{\alpha}V}{NQ} \int_0^1 \mathrm{d}sq(\mathbf{r}, s)q'(\mathbf{r}, 1-s)\delta_{\alpha, \tau(s)}$$
(S2)

where $\tau(s)$ describes the monomer sequence on chain. $q(\mathbf{r}, s)$ and $q'(\mathbf{r}, s)$ are partial partition functions that satisfy the diffusion equation

$$\frac{\partial q\left(\mathbf{r},s\right)}{\partial s} = R_g^2 \nabla^2 q\left(\mathbf{r},s\right) - \omega\left(\mathbf{r}\right) q\left(\mathbf{r},s\right)$$
(S3)

with initial condition $q(\mathbf{r}, 0) = q'(\mathbf{r}, 0) = 1$ and $\omega(\mathbf{r}) = \omega_{\rm A}(\mathbf{r})$ or $\omega_{\rm B}(\mathbf{r})$ based on the parameter

s. $q(\mathbf{r}, s)$ is obtained by setting $\omega(\mathbf{r}) = \omega_{\rm A}(\mathbf{r})$ for $s < N_{\rm A}/N$ and $\omega(\mathbf{r}) = \omega_{\rm B}(\mathbf{r})$ otherwise, Conversely, $q'(\mathbf{r}, s)$ is defined by setting $\omega(\mathbf{r}) = \omega_{\rm B}(\mathbf{r})$ for $s < N_{\rm B}/N$ and $\omega(\mathbf{r}) = \omega_{\rm A}(\mathbf{r})$ otherwise.

The calculation of the single chain partition function Q is then accomplished by

$$Q = \frac{1}{V} \int d\mathbf{r} q(\mathbf{r}, 1) = \frac{1}{V} \int d\mathbf{r} q'(\mathbf{r}, 1)$$
(S4)

More details and the derivations of above equations can be found, e.g. in refs 1, 4 and 5.

At equilibrium, the free energy functional $F\{\phi_{\alpha}(\mathbf{r})\}$ reaches its minimum with respect to the density fields $\phi_{\alpha}(\mathbf{r})$, leading to a second set of conditions for the values of the conjugate fields ω_{α}

$$\omega_{\rm A}^{\rm SCF}(\mathbf{r}) = \chi N \phi_{\rm B}$$

$$\omega_{\rm B}^{\rm SCF}(\mathbf{r}) = \chi N \phi_{\rm A}$$
(S5)

However, in DDFT calculations, these conditions are not imposed. Instead, the system is dynamically driven toward the equilibrium state via the diffusive dynamical eq. 2 with $\mu(\mathbf{r}) = \mu_{\rm A}(\mathbf{r}) - \mu_{\rm B}(\mathbf{r}) = (\omega_{\rm A}^{\rm SCF}(\mathbf{r}) - \omega_{\rm A}(\mathbf{r})) - (\omega_{\rm B}^{\rm SCF}(\mathbf{r}) - \omega_{\rm B}(\mathbf{r})).$

In the present work, the SCFT calculations are effectively one-dimensional, that is, we assume that densities vary only in the z-direction. And, the conjugate fields, eq. S2. is solved using the pseudo spectral scheme² with discrimination $\Delta s = 0.01$.

S2 Dynamic Density Functional Theory

S2.1 Schemes for Onsager coefficient

For melts in the Rouse regime (i.e., non-entangled chains), four types of Ansatz for the Onsager coefficient $\Lambda(\mathbf{r}, \mathbf{r}')$ in eq. 1 have been proposed in the literatures:

(i) Local Coupling Scheme: Here, monomer beads are assumed to diffuse independently of each other with the mobility D_0 .⁶ This leads to the following expression for $\Lambda_{\alpha\beta}(\mathbf{r},\mathbf{r}')$

$$\Lambda_{\alpha\beta}^{\text{Local}}(\mathbf{r},\mathbf{r}') = D_0 \phi_\alpha(\mathbf{r}) \delta_{\alpha\beta} \delta(|\mathbf{r}-\mathbf{r}'|)$$
(S6)

Thus, for our system, the Onsager coefficient of local scheme in eq. 2 is

$$\Lambda^{\text{Local}}(|\mathbf{r} - \mathbf{r}'|) = \frac{1}{2} D_0 \delta(|\mathbf{r} - \mathbf{r}'|)$$
(S7)

(ii) Chain Coupling Schemes: They assume that the polymer chains diffuse as a whole with mobility, $D_c = D_0/N$. For this case, Maurits and Fraaije have derived the expression⁷

$$\Lambda_{\alpha\beta}^{\text{Chain}}(\mathbf{r}, \mathbf{r}', t) = D_c P_{\alpha\beta}(\mathbf{r}, \mathbf{r}', t)$$
(S8)

where $P_{\alpha\beta}(\mathbf{r}, \mathbf{r}', t)$ is the pair correlation of monomers α and β on the same chain at position \mathbf{r} and \mathbf{r}' . $P_{\alpha\beta}$ is further approximated by the pair correlations of ideal Gaussian chains, known as the Debye correlation function⁸

$$\Lambda_{\alpha\beta}^{\text{Debye}}(\mathbf{r},\mathbf{r}') = D_{c}g_{\alpha\beta}(|\mathbf{r}-\mathbf{r}'|)$$
(S9)

Analytical expressions of $g_{\alpha\beta}(|\mathbf{r} - \mathbf{r}'|)$ are available for the Fourier representation $g_{\alpha\beta}(\mathbf{q})$. For example, for diblock copolymers one obtains^{2,9}

$$g_{aa}(\mathbf{q}) = Ng_{\rm D}(f_a, x)$$

$$g_{\rm AB} = \frac{N}{2} \{ g_{\rm D}(1, x) - g_{\rm D}(f_{\rm A}, x) - g_{\rm D}(f_{\rm B}, x) \}$$
(S10)

where $x = \mathbf{q}^2 R_g$, f_{α} is the volume fraction of block α , and $g_{\mathrm{D}}(f, x) = \frac{2}{x^2}(fx + e^{-fx} - 1)$ is the well-known Debye function.

Consequently, the Onsager coefficient of Debye scheme in eq 3 under our system is

$$\Lambda^{\text{Debye}}(\mathbf{q}) = 2ND_c g_{\text{D}}(\frac{1}{2}, x) - \frac{1}{2}ND_c g_{\text{D}}(1, x)$$
(S11)

(iii) Mixed Coupling Schemes: The prediction of local and nonlocal schemes have shown to underestimate and overestimate the ordering time, respectively, when compared to simulations.^{10,11} Thus, the mixed scheme is proposed,¹¹ which assumes simultaneously dynamics governed by a local mobility function at short wavelengths and a nonlocal one at large wavelengths, by introducing a filter function $\Xi(\mathbf{r})$ to filter out the long-wavelength part of the thermodynamic driving force

$$f_{\alpha}^{\text{Nonlocal}}(\mathbf{r}) = -\int d\mathbf{r}' \Xi(|\mathbf{r} - \mathbf{r}'|) \nabla \mu_{\alpha}(\mathbf{r}')$$
(S12)

with

$$\Xi(r) = (2\pi\xi^2)^{-3/2} \exp\left\{\frac{-r^2}{2\xi^2}\right\}$$
(S13)

Therefore, the remaining part is short wavelengths of force that drive local rearrangements of the chain.

$$f_{\alpha}^{\text{Local}}(\mathbf{r}) = -\nabla \mu_{\alpha}(\mathbf{r}) - f_{\alpha}^{\text{Nonlocal}}(\mathbf{r})$$
(S14)

The resulting mixed scheme has the form

$$\frac{\partial \phi_{\alpha}(\mathbf{r},t)}{\partial t} = -\nabla \sum_{\beta} \int d\mathbf{r}' \left[\Lambda_{\alpha\beta}^{\text{Nonlocal}}(\mathbf{r},\mathbf{r}') \mathbf{f}_{\beta}^{\text{Nonlocal}}(\mathbf{r}') + \Lambda_{\alpha\beta}^{\text{Local}}(\mathbf{r},\mathbf{r}') \mathbf{f}_{\beta}^{\text{Local}}(\mathbf{r}') \right]$$
(S15)

where $\Lambda^{\text{Nonlocal}}$ can be any of the chain coupling schemes discussed above. In the main text, we use Debye scheme as a nonlocal mobility function as in ref 11.

Then, we can conveniently rewrite the mixed scheme (eq. S15) in the form of eq. 1

$$\frac{\partial \phi_{\alpha}(\mathbf{r},t)}{\partial t} = \nabla \left\{ \int d\mathbf{r}' \left(\delta(|\mathbf{r}-\mathbf{r}'|) - \Xi \left(|\mathbf{r}-\mathbf{r}'|\right) \right) \phi_{\alpha} \nabla \mu_{\alpha}(\mathbf{r}',t) + \sum_{\beta} \int d\mathbf{r}' G_{\alpha\beta}(|\mathbf{r}-\mathbf{r}'|) \nabla \mu_{\beta}(\mathbf{r}',t) \right\}$$
(S16)

with

$$G_{\alpha\beta}(|\mathbf{r} - \mathbf{r}'|) = \int d\mathbf{r}'' g_{\alpha\beta} \left(|\mathbf{r} - \mathbf{r}''| \right) \Xi \left(|\mathbf{r}'' - \mathbf{r}'| \right)$$
(S17)

Therefore, for our incompressibility system, the evolution of the density ϕ can be calculated under the form of eq. 2 by the following equation

$$\frac{\partial \phi(\mathbf{r}, t)}{\partial t} = \nabla \int d\mathbf{r}' \Lambda^{\text{Mixed}}(|\mathbf{r} - \mathbf{r}'|) \nabla \mu(\mathbf{r}', t)$$
(S18)

with

$$\Lambda^{\text{Mixed}}(|\mathbf{r} - \mathbf{r}'|) = G_{\text{AA}}(|\mathbf{r} - \mathbf{r}'|) - G_{\text{AB}}(|\mathbf{r} - \mathbf{r}'|) + \frac{1}{2}\delta(|\mathbf{r} - \mathbf{r}'|) - \frac{1}{2}\Xi(|\mathbf{r} - \mathbf{r}'|)$$
(S19)

The tunable parameter ξ in $\Xi(|\mathbf{r} - \mathbf{r}'|)$ term determines the length scale of crossover between the local and the Debye dynamics. Figure S1 illustrates the impact of the tunable parameter ξ on Λ^{Mixed} in Fourier space. When $\xi = 0$, $\Lambda^{\text{Mixed}} = \Lambda^{\text{Debye}}$, and as $\xi \to \infty$, $\Lambda^{\text{Mixed}} = \Lambda^{\text{Local}}$. In the main text, the tunable parameter is set to $\xi = 0.5R_g$, as optimized in previous work.¹¹

(iv) Relaxation Time Scheme: Mantha et al.¹² propose to estimate Onsager coefficient



Figure S1: Influence of the tunable parameter ξ on $\Lambda^{\rm Mixed}$ in Fourier space

directly from the characteristic relaxation time of the single chain dynamic structure factor $g(\mathbf{q},t) = \frac{1}{N} \langle \sum_{m,j=1}^{N} e^{i\mathbf{q} \cdot (\mathbf{R}_m(t) - \mathbf{R}_j(0))} \rangle$, where $\mathbf{R}_j(t)$ is the position of monomer j at time t. Knowing $g(\mathbf{q},t)$, the rescaled single-chain mobility in Fourier space is

$$\Lambda^{\text{RTA}}(\mathbf{q}) = \frac{1}{k_{\mathbf{B}}Tq^2N}G^{-1}(\mathbf{q})g(\mathbf{q},0) \text{ with}$$

$$G(\mathbf{q}) = \int_0^\infty \mathrm{d}t g(\mathbf{q},t)g^{-1}(\mathbf{q},0)$$
(S20)

In the case of symmetric diblock copolymers with fully equivalent A and B blocks, the Onsager coefficient is then given by

$$\Lambda_{\rm AA}^{\rm RTA}(\mathbf{q}) = \frac{1}{4k_{\rm B}Tq^2N} \left(\frac{g(\mathbf{q},0)}{\tau_{\rm R}} + \frac{\Delta(\mathbf{q},0)}{\tau_{\Delta}} \right)$$
$$\Lambda_{\rm AB}^{\rm RTA}(\mathbf{q}) = \frac{1}{4k_{\rm B}Tq^2N} \left(\frac{g(\mathbf{q},0)}{\tau_{\rm R}} - \frac{\Delta(\mathbf{q},0)}{\tau_{\Delta}} \right)$$
(S21)

where $\Delta(\mathbf{q},t) = g_{AA}(\mathbf{q},t) + g_{BB}(\mathbf{q},t) - g_{AB}(\mathbf{q},t) - g_{BA}(\mathbf{q},t)$ and $\tau_{\Delta} = \frac{1}{\Delta(\mathbf{q},0)} \int_{0}^{\infty} \mathrm{d}t \Delta(\mathbf{q},t)$.

Then, for our system, the Onsager coefficient of relaxation time scheme is

$$\Lambda^{\rm RTA}(\mathbf{q}) = \frac{2\Delta(\mathbf{q},0)}{4k_{\rm B}Tq^2N\tau_{\Delta}} \tag{S22}$$

where the structure factor $g(\mathbf{q}, t)$ is obtained through Brownian dynamics at $\chi N = 0.^{12}$

S2.2 Settings of DDFT calculation

In practice, the DDFT calculations are carried out in one dimension, since we are only concerned with the lamellar morphology in the z direction. As in earlier work,^{11,12} the time step depends on the DDFT scheme. We use $dt = 10^{-5}\tau$ for the local dynamics model, $dt = 10^{-4}\tau$ for the Debye and relaxation time dynamics models, and $dt = 10^{-6}\tau$ for the mixed dynamics model.

S2.3 Influence of the absence of wavenumber $|\mathbf{q}| < 2$ in $\Lambda(\mathbf{q}, t)$ on $\Lambda(|z - z'|, t)$

For our system, the $\Lambda^{ML}(\mathbf{q}, t)$, obtained by machine learning, actually lacks the values for wavenumbers $|\mathbf{q}| < 2$ in Fourier space. In this section, we will demonstrate through testing with Λ^{Debye} that the absence of values for $|\mathbf{q}| < 2$ does not significantly affect the Onsager coefficient in real space.

To fully illustrate this, we respectively selected Λ^{Debye} (Figure S2a), where $\Lambda^{\text{Debye}}(q = 0) = 0$, and $\Lambda^{\text{Debye}}_{AA}$ (Figure S2c), where $\Lambda^{\text{Debye}}_{AA}(q = 0) \neq 0$, for testing by varying the box size L_z . The box size $L_z = 3R_g$ was used in the main text. As L_z increases, the spacing of q decreases, resulting in multiple values in $|\mathbf{q}| < 2$. However, for both Λ^{Debye} (Figure S2b) and $\Lambda^{\text{Debye}}_{AA}$ (Figure S2d), there is no significant change for the Onsager coefficient in real space.



Figure S2: Influence of the spacing of q in Fourier space (a, c) by changing the box size L_z on the Onsager coefficients Λ^{Debye} (b) and $\Lambda^{\text{Debye}}_{\text{AA}}$ (d) in real space.

S3 Brownian Dynamics Simulations

S3.1 Simulation method

In this section, We briefly describe the Brownian dynamics (BD) simulations, referred to as *fine-grained* simulation. Due to our aim of extracting parameters from particle-based models to reproduce the dynamical properties on the basis of the field-based model, we employ an Edwards-type model, ^{13–15} whose static equilibrium properties are well-reproduced by density functional theory with minimal parameter adjustment.¹¹

In our study, we consider melts comprising $n_c = 5000$ block copolymers, each with a chain length N = 40, containing $N_A = 20$ beads of type A and $N_B = 20$ beads of type B, with mobility $D_c = D_0/N$ in a box of volume $V = L_x \cdot L_y \cdot L_z = R_g \times R_g \times 3R_g$ with periodic boundary. The average monomer density is thus $\rho_0 = n_c N/V$.

The interactions of the Edwards-type model, expressed in terms of local densities,¹⁵ are described by the Hamiltonian H_{energy} , which is given by¹¹

$$H_{\text{energy}} = \frac{N}{4} \sum_{m=1}^{n_c} \sum_{j=1}^{N} \left(\mathbf{R}_{m,j} - \mathbf{R}_{m,j-1} \right)^2 + \rho_0 \chi \int d\mathbf{r} \hat{\phi}_A(\mathbf{r}) \hat{\phi}_B(\mathbf{r}) + \rho_0 \kappa \int d\mathbf{r} \left(\hat{\phi}_A(\mathbf{r}) + \hat{\phi}_B(\mathbf{r}) - 1 \right)^2$$
(S23)

The first term represents the bonded interactions within the polymer, while the subsequent terms correspond to nonbonded interactions with the Flory-Huggins interaction parameter χ , which controls the incompatibility between A and B monomers, and the Helfand parameter κ , which controls the compressibility. The $\mathbf{R}_{m,j}$ is the position of the *j*-th bead of *m*-th chain, and the $\hat{\phi}_{\alpha}(\mathbf{r})$ is the normalized microscopic densities of α -type beads ($\alpha = A$ or B) at position \mathbf{r} , defined as,

$$\hat{\phi}_{\alpha}(\mathbf{r}) = \frac{1}{\rho_0} \sum_{mj} \delta(\mathbf{r} - \mathbf{R}_{mj}) \delta_{\alpha, \tau_{mj}}$$
(S24)

where $\tau_{mj} = A$ or B characterizes the monomer sequence on chain m. In practice, the local densities are evaluated on a grid of size $\Delta x = \Delta y = 0.1R_g$ and $\Delta z = 0.0468R_g$, using a first-order cloud-in-cell (CIC) scheme,¹⁶ where beads influence the densities at the nearest eight mesh points. Further insights on the CIC method in BD simulations can be found in ref. 11.

The motion of a BD bead is governed by deterministic conservative forces derived from the Hamiltonian, as well as stochastic forces. These forces are mathematically expressed as:

$$\frac{\mathrm{d}\mathbf{R}_{m,j}}{\mathrm{d}t} = -D_0 \frac{\partial H_{\mathrm{energy}}}{\partial \mathbf{R}_{m,j}} + \sqrt{2D_0} \mathbf{f}_{m,j} \tag{S25}$$

where the random force $\mathbf{f}_{m,j}$ is Gaussian distributed random noise with zero mean and variance $\langle f_{mjI}(t) f_{nkJ}(t') \rangle = \delta_{mn} \delta_{jk} \delta_{IJ} \delta(t-t')$, where I, J denote the Cartesian components. The BD equation is a stochastic differential equation, and it is integrated using the explicit Euler-Maruyama method¹⁷ with a time step of $dt_{\rm BD} = 0.001 \tau/N$ (i.e. $dt_{\rm BD} = 2.5 \times 10^{-5}$ at N = 40). For a detailed theory and setup of Brownian dynamics simulations, we refer to refs. 11 and 12.

In particle, the Flory-Huggins interaction parameter is set to $\chi N = 12 \sim 20$ and the Helfand parameter is set to $\kappa N = 10$. And we initialize the system by imposing weakly inhomogeneous density distributions and monitor the evolution of the density profile ϕ until it is equilibrated.

S3.2 Influence of compressibility parameter on density

Figure S3 shows the results of BD simulations at different κN . When κN is within a relatively small range, the density is not significantly affected by κN . However, high $\kappa N \geq 500$, the interactions in the BD simulations are significantly altered, preventing the formation of lamellar morphology. Therefore, despite assuming incompressibility in our main text, we still generated data using $\kappa N = 10$ and then used eq. 4 in main text to enforce density incompressibility.



Figure S3: Results of BD simulations with different compressibility parameter κN by using *cloud in cell* method. (a) The time evolution of the maximum density of A-monomers ϕ_A^{max} . (b) Density profiles ϕ_{α} of A-monomers and B-monomers at equilibrium. (c) Total Density profiles $\phi_A + \phi_B$ at equilibrium

In addition, the evolution of density profiles at $\chi N = 12$ and $\kappa N = 10$ with the *cloud in cell* method is shown in Figure S4.



Figure S4: Evolution of density profiles $\phi(z,t)$ at $\chi N = 12$ and $\kappa N = 10$ with *cloud in cell* method.

S4 Coarse-grained Density Fields

S4.1 Kernel coarse-graining with periodic boundaries

To coarse-grain the discrete microscopic data through main text eq. 4, we used a 3D Gaussian kernel

$$K(x, y, z) = \frac{1}{(2\pi)^{\frac{3}{2}}\sigma^3} \exp\left(-\frac{x^2 + y^2 + z^2}{2\sigma^2}\right)$$
(S26)

Meanwhile, periodicity of the coarse-grained fields for the microscopic data was ensured by placing ghost particles periodically around the domain.

S4.2 Independence of Gaussian kernel width σ on the density fields

To demonstrate that the obtained density fields are not influenced by the Gaussian kernel width σ in the *particle-to-field convolution with a kernel* method (section *Particle-to-field transformations* in the main text), we selected several similar σ values within the plateau $H(\sigma)$. Following the steps outlined in section *Particle-to-field transformations* of the main text, we obtained the density fields, shown in Figure S5, at $\chi N = 12$. Our results indicate that the resulting density fields are unaffected by the choice of Gaussian kernel width σ .



Figure S5: Evolution of density fields $\phi(z, t)$ with different Gaussian kernel width σ in the plateau $H(\sigma)$ at different time.

S5 Machine Learning Methods

S5.1 Architecture of the DDFT-Informed ODE Net

The DDFT-Informed ODE Net (DIO Net), inspired by DDFT, comprises two sub-networks, i.e., ODE net and Neural Net, which in general can have different structures (see Figure 1).

In the ODE Net, There are n_q small networks, each of which extracts the time partial derivative of the density under different Fourier bases, $\partial_t \phi^{\text{ML}}(\mathbf{q}, t)$, due to the independence of density evolution with respect to \mathbf{q} as implied by eq. 3 in the main text. In our model, n_q is chosen from 8 to 12, and it has been verified that increasing n_q beyond this range does not impact our results. Each of these small networks consists of two hidden layers, each containing 5 nodes activated by ELU functions, in addition to single-node input and output layers. We have verified that the machine learning results remain consistent when using different activation functions, such as a tanh function. It is important to note that these $n_{\mathbf{q}}$ small networks in real space to obtain the time partial derivative of the density. However, in practice, we find that using networks with a small number of free parameters in Fourier space significantly enhances training efficiency without sacrificing model compatibility and predictive power. In general, the number of free parameters can be minimized by considering the properties of the physical system, while networks with redundant parameters can work equally well if proper regularization is imposed.¹⁸

Simultaneously, another Neural Net, consisting of an input layer and an output layer with a single node, as well as two hidden layers containing 50 nodes each, is utilized to learn the Onsager coefficient $\Lambda^{ML}(\mathbf{q})$. Both the time partial derivative of the density $\partial_t \phi^{ML}(\mathbf{q}, t)$ and the Onsager coefficient $\Lambda^{ML}(\mathbf{q})$ are transformed into real space via fast Fourier transforms and are coupled through the DDFT equations (eq. 2 in main text).

S5.2 Training the DDFT-Informed ODE Net

We use the ODE Net to find the time partial derivative of the density by employing the adjoint sensitivity method.¹⁹ This involves the minimization of the error, which measures the accuracy of the learned dynamics, at discrete-time points t_{i+j} with $j = 1, 2, ..., n_t$ and at an arbitrary time $t_i \in [t_0, t_e]$, between the ODE Net predictions $\hat{\phi}(\mathbf{r}, t)$ and the set of observation data $\phi(\mathbf{r}, t)$ using e.g., the mean absolute error,

$$loss_{ODE} = \frac{1}{N_t n_t} \sum_{i=1}^{N_t} \sum_{j=1}^{n_t} \left(\left| \hat{\phi}_{i,j}(\mathbf{r}, t) - \phi_{i,j}(\mathbf{r}, t) \right| \right)$$
(S27)

The selection of n_t may influence the final outcomes. When n_t is very small, especially when $n_t = 1$, due to the small density variation and the high noise in particle simulation, the model may result in a trivial solution $\partial_t \phi^{\text{ML}}(\mathbf{r}, t) = 0.20$ Conversely, when n_t is very large, especially when $n_t = N_t$, it substantially slows down the model training process.^{19,20} Therefore, in this paper, we choose $n_t \approx N_t/6$, which balances the training performance and ensures high accuracy.

The neural network extracts the Onsager coefficient $\Lambda(|\mathbf{r} - \mathbf{r}'|)$ from the time partial derivative of the density by minimizing the loss of the DDFT equations,

$$loss_{DDFT} = \frac{1}{N_t} \sum_{i=1}^{N_t} \left| \partial_t \hat{\phi}(\mathbf{r}, t_i) - \nabla \int d\mathbf{r}' \Lambda(|\mathbf{r} - \mathbf{r}'|) \nabla \mu(\mathbf{r}, t_i) \right|$$
(S28)

where $\mu(\mathbf{r}, t)$ is the chemical potential, obtained using the SCFT method¹ with $\phi(\mathbf{r}, t)$.

The total loss is then

$$loss = loss_{ODE} + \eta loss_{DDFT} \tag{S29}$$

where η is a parameter to balance the accuracy of the learned dynamics and the error from DDFT. In our computation, we first train the loss function with $\eta = 0$ to obtain the time partial derivative of the density. After some training steps, we set $\eta = 1$ to obtain the Onsager coefficient and fine-tune the parameters of the ODE Net, since both losses are equally important and are of the same order of magnitude (see Figure S6). In the future, more schemes of adaptive loss balancing can be explored.^{21,22}

We use the Adam optimizer²³ for updating the network parameters. The learning rate was set to decay by 0.8 if the *loss* did not decrease within 2,000 epochs, starting from an initial value of 0.001, to obtain the optimal Onsager coefficient. Figure S6 shows the best *loss*, *loss*_{ODE}, *loss*_{DDFT}, and their corresponding learning rates over the training epochs for the phase with $\eta = 1$. All code of the ML methods was implemented in python with the PyTorch package.²⁴



Figure S6: Learning curves as a function of the number of epochs for training data at $\chi N = 12$. The black line indicates the best *loss*, which combines the *loss*_{ODE} (blue line) and *loss*_{DDFT} (cyan line). The red line represents the corresponding learning rate (lr).

S5.3 Making predictions using Λ^{ML}

Once well trained, we can obtain the Onsager coefficient $\Lambda^{ML}(\mathbf{r}, \mathbf{r}')$ from the DDFT-Informed ODE Net. We incorporate the obtained $\Lambda^{ML}(\mathbf{r}, \mathbf{r}')$ into the DDFT equations and evolve from initial time t_0 , which allows us to obtain the evolution of density fields under this $\Lambda^{ML}(\mathbf{r}, \mathbf{r}')$. The time evolution of the maximum density for different χN is shown in the Figure S7, demonstrating that our obtained $\Lambda^{ML}(\mathbf{r}, \mathbf{r}')$ can reproduce the density evolution process observed in particle simulations.



Figure S7: The time evolution of the maximum density ϕ^{max} with different incompatibility parameter χN as obtained from BD simulations and the DDFT models with Λ^{ML} .

S5.4 Additional results for numerical fitting

In addition to fitting Onsager coefficient at $\chi N = 12$ using the stretched exponential function employed in the main text, we also attempted to fit it using other functions, as shown in Figure S8. The first function we tried was exponential function $\Lambda(\Delta z) = a_0 \exp\left(-\frac{\Delta z}{r_0}\right)$, which yielded poor fitting results. Additionally, we used double stretched exponential function, $\Lambda(\Delta z) = a_1 \exp\left(-(\frac{\Delta z}{r_1})^{\beta_1}\right) + a_2 \exp\left(-(\frac{\Delta z}{r_2})^{\beta_2}\right)$, for fitting and found that the r_1 is similar to r_2 , further demonstrating that a single stretched exponential function is sufficient to describe Onsager coefficient.

In addition, the fitting parameter values of $\Lambda(\Delta z) = a_0 \exp\left(-\left(\frac{\Delta z}{r_0}\right)^{\beta_0}\right)$ at different χN are shown in Table S1.

S5.5 Speedup factor of DDFT simulations

Since our method can extract Onsager coefficients Λ from particle-based BD simulations to reproduce the dynamic properties on the basis of field-based model, field-based models can



Figure S8: Numerical fitting of the learned Onsager coefficient Λ^{ML} at $\chi N = 12$ in real space with different equations. The parameters of $\Lambda(\Delta z) = a_1 \exp\left(-\left(\frac{\Delta z}{r_1}\right)^{\beta_1}\right) + a_2 \exp\left(-\left(\frac{\Delta z}{r_2}\right)^{\beta_2}\right)$ are $r_1 = 0.817$, $r_2 = 0.868$, $\beta_1 = 1.679$ and $\beta_2 = -2.085$, where $\Delta z = |z - z'|$.

Table S1: Numerical fitting results of the learned Onsager coefficient in real space by using $\Lambda(\Delta z) = a_0 \exp\left(-\left(\frac{\Delta z}{r_0}\right)^{\beta_0}\right)$

χN	r_0	β_0
12	0.824(9)	1.74(3)
12.4	0.749(4)	1.82(2)
13	0.668(6)	1.84(4)
14	0.646(9)	1.51(4)
15	0.629(2)	1.87(2)
17	0.574(8)	1.63(5)
20	0.497(4)	1.76(3)

be directly used to simulate the evolution of density, thereby avoiding the time-consuming BD simulation. Thus, our method can be used for simulation acceleration.

Figure S9 depicts the speedup ratios at different time steps for DDFT, dt_{DDFT} . When using the same time step as the BD simulation, i.e., $dt_{\text{DDFT}} = 0.001/N\tau$, our current system achieves nearly a 20-fold acceleration. Interestingly, as the time steps dt_{DDFT} increase, the speedup factor further increases, reaching up to nearly 450 times at $dt_{\text{DDFT}} = 0.01\tau$, while the dynamic evolution remains consistent with the particle simulation (Figure S9a). Even with a time step smaller than that of the particle simulation, i.e., $dt_{\text{DDFT}} = 10^{-5}\tau$, it retains an acceleration ratio of nearly 12 times.

Therefore, compared to particle-based simulations, field-based models with reasonable parameters not only reproduce the dynamic properties of particle simulations but also offer significantly higher acceleration.



Figure S9: (a) The time evolution of the maximum density ϕ^{max} with different time steps for DDFT, dt_{DDFT} . (b) Speedup factor of DDFT simulations at different time steps dt_{DDFT} compared to BD simulations with $dt_{\text{BD}} = 0.001/N\tau$.

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