Morphological characterization of bicontinuous microstructures using two-point statistics

Y Sun, K L M Elder and P W Voorhees
Department of Materials Science and Engineering, Northwestern University, Evanston IL 60208
E-mail: p-voorhees@northwestern.edu

Abstract. Bicontinuous structures are found in a wide range of multiphase materials. We employ the prototypical bicontinuous structure that is found following spinodal decomposition to probe the morphology and evolution of bicontinuous two-phase mixtures during coarsening using two-point statistics of the mean, total curvature, and the interfacial velocity. We find that the relative spatial variations of the interfacial velocity are larger than those of the mean curvature. In addition, interfaces with high positive and high negative values of the mean curvature, total curvature and interfacial velocity are spatially localized. Finally, the spatial distribution of interfaces with near zero interfacial velocity largely resembles that of near zero mean curvature, and have correlations only over very short distances.

1. Introduction
Bicontinuous microstructures usually form as a result of phase ordering or phase decomposition such as spinodal decomposition. These microstructures have complex morphologies and topologies, and have been observed in various types of materials in nature, including metals, polymers, and ceramics. These bicontinuous structures are a result of phase decomposition which occurs when a homogeneous single-phase mixture is cooled into a two-phase region and the alloy phase separates. After the initial phase decomposition, the system may continue to evolve towards a lower total energy by reducing the total interfacial area, in other words, the system will undergo coarsening. During the coarsening processes, the amount of each phase remains constant throughout the evolution process. In this case, the system evolves under conserved dynamics. In other cases, such as spin ordering in magnetic materials, a region in the system may transition from one phase to the other without being compensated by an opposite transition somewhere else in the system. This type evolution is said to follow non-conserved dynamics.

Although it has been shown that dendritic microstructures do not show self-similarity during coarsening, studies have shown that bicontinuous microstructures do evolve in a self-similar manner during coarsening. Jinnai et. al. examined the time-evolution of the probability distributions of the mean and Gaussian curvatures in a binary polymer mixture formed via spinodal decomposition. They showed that the probability distributions of the mean and Gaussian curvatures, when scaled by $S_v(t)$ and $S_v(t)$, respectively, where $S_v$ is the surface area per volume and $t$ is time, become independent of time in the evolution. This implies that the microstructure evolves in a self-similar manner. In other investigations the morphological evolution of simulated bicontinuous structures where the morphology
is characterized using interfacial shape distributions (ISDs) was studied. The ISD is the probability of finding a patch of interface with a given pair of principle curvatures $\kappa_1, \kappa_2$. It was demonstrated that, for both bicontinuous structures simulated via Cahn-Hilliard (conserved) [11] and Allen-Cahn (non-conserved) [12] dynamics, the scaled morphology remains constant during the late stage of coarsening. This again supports the self-similarity of the coarsening process in bicontinuous microstructures. During coarsening of these structures under conserved dynamics, the characteristic length scale, given by $S^{-1}$, follows the expected $t^{1/3}$ power law [13]. This temporal scaling law of $S^{-1}$ applies to bicontinuous structures regardless of the phase volume fractions of the phases.

While $S^{-1}$ can be used as a descriptor of the characteristic length scale of the bicontinuous microstructure during coarsening which follows a temporal power law, using $S^{-1}$ alone is far from sufficient to describe the complicated morphology of the bicontinuous microstructure during coarsening. Although the ISD, similar to a probability distribution of a polydisperse array of spheres, is an important measure of the interfacial morphology, it does not give any insights into how these patches of curvature are distributed in the structure. To address the spatial distribution of curvature in the structure and further characterize these complex structures, we shall investigate two-point statistics of the interfacial curvature.

2. Two-point statistics introduction

We split the interface, $\Omega$, into small patches at spatial locations denoted by $x$. Interfacial properties (interfacial curvatures $H, K, d$, and normal velocity $V$, where the mean curvature $H = (\kappa_1 + \kappa_2)/2$, the Gaussian curvature $K = \kappa_1 \kappa_2$, and the total curvature $d = \sqrt{\kappa_1^2 + \kappa_2^2}$) are calculated at these locations, which constitute a set of microstructure functions as a function of $x$ denoted by $m(u, x)$ with $u$ being the interfacial property considered. In practice, we will have a set of microstructure functions $m(H, x), m(K, x), m(V, x)$, etc. defined at each interface patch at $x \in \Omega$. We then calculate the two-point Pearson correlation of these microstructure functions. The two-point Pearson correlation of microstructure functions associated with interfacial properties $u$ and $u'$ is defined as

$$g(u, u' | r) = \frac{1}{|\Omega(r)|} \frac{1}{\sigma(r)\sigma'(r)} \int_{x \in \Omega(r)} [m(u, x) - \mu] [m(u', x + r) - \mu'] \, dx,$$  \hspace{1cm} (1)

where $\mu$ and $\mu'$ are the mean values of $m(u, x)$ and $m(u', x)$ on the interface $\Omega$, $\sigma(r)$ and $\sigma'(r)$ are the standard deviations, given by

$$\sigma(r) = \left\{ \frac{1}{|\Omega(r)|} \int_{x \in \Omega(r)} [m(u, x) - \mu]^2 \, dx \right\}^{\frac{1}{2}},$$  \hspace{1cm} (2)

$$\sigma'(r) = \left\{ \frac{1}{|\Omega(r)|} \int_{x \in \Omega(r)} [m(u', x + r) - \mu']^2 \, dx \right\}^{\frac{1}{2}},$$  \hspace{1cm} (3)

and $\Omega(r)$ is the set of all interfacial patches (locations) where both $m(u, x)$ and $m(u', x + r)$ are defined, and $|\Omega(r)|$ is the total interfacial area represented by $\Omega(r)$.

Although this function is fully capable of mapping over continuous spaces, any practical implementation on either experimental or simulated datasets demands the adoption of one of the many discretized variants of this function. It is also noted that the discretized versions are better suited for computer programming. We examine next the details of these discretized variants.

It is assumed that the microstructure volume of interest occupies a rectangular parallelepiped space $\mathbb{X}$ that can be uniformly tessellated into spatial bins (i.e., voxels) $X_s \in \mathbb{X}$, where
s = 1, 2, ... is the integer index enumerating the spatial bins. Note that s may be a vector in a high-dimensional space. In other words, for a three-dimensional volume, one might use a vector s = (s1, s2, s3) to index each spatial bin Xs in the microstructure volume, where s1, s2, and s3 take integer values. In a similar way, we could tessellate the local state space U in some suitable manner to identify discretized bins in the local state space. In other words, we can discretize U into bins Un ∈ U, with n = 1, 2, ... (could also be a vector index if needed) enumerating these local state bins. Building on the discretized representations, we can now invoke a discretized representation for the microstructure function as m[n, s], which denotes the volume fraction in the spatial bin Xs occupied by local states identified by Un. Specifically,

\[ m[n, s] = \int_{u \in U_n} \int_{x \in X_s} m(u, x) \, dx \, du. \]  

(4)

With this definition, it is easy to see that, at each spatial bin Xs, the volume fractions of all local states Un have values between 0 and 1, and they sum to 1,

\[ \sum_n m[n, s] = 1, \quad 0 \leq m[n, s] \leq 1 \forall n. \]  

(5)

When the discretization in space is fine enough, each spatial bin Xs will only contain local states belonging to one local state bin Un. In that case, the microstructure function m[n, s] will be a binary function taking values of 0 or 1,

\[ m[n, s] = \begin{cases} 
1, & u(x) \in U_n \forall x \in X_s \\
0, & \text{otherwise} 
\end{cases} \]  

(6)

It should be noted that the binning of the local state space occurs quite naturally in some situations. For example, if one is dealing with a multiphase material system, then each phase can be simply assigned a different value of n. In particular, in a two-phase system, N could take on values of 0 and 1.

Following the approaches described earlier, it is relatively straightforward to write discrete versions of both two-point Pearson correlations introduced above. In this work, because the local states of interest exhibit values on a continuous range, we are primarily interested in the application of the two-point Pearson correlations based directly on the values of the local states (e.g., interface curvature and interface velocity). Therefore, the corresponding discretized versions are

\[ \tilde{g}[u, u’ | t] = \frac{1}{|S[t]|} \frac{1}{\tilde{a}[t] \tilde{a’}[t]} \sum_{s \in S} \left( u[s] - \tilde{\mu} \right) c[s] \cdot \left( u’[s + t] - \tilde{\mu’} \right) c[s + t], \]  

(7)

\[ \tilde{\mu} = \frac{1}{|S[0]|} \sum_{s \in S} u[s] c[s], \]  

(8)

\[ \tilde{\mu’} = \frac{1}{|S[0]|} \sum_{s \in S} u’[s] c[s], \]  

(9)

\[ \tilde{\sigma}[t] = \left\{ \frac{1}{|S[t]|} \sum_{s \in S} \left| u[s] - \tilde{\mu} \right|^2 c[s] c[s + t] \right\}^{\frac{1}{2}}, \]  

(10)
\[ \bar{\sigma}'[t] = \left\{ \frac{1}{|S[t]|} \sum_{s \in S} \left| u'[s + t] - \bar{\mu}' \right|^2 c[s] c[s + t] \right\}^{\frac{1}{2}}, \quad (11) \]

where
\[ |S[t]| = \sum_{s \in S} c[s] c[s + t]. \quad (12) \]

However, we will also use the discretized local states to aid our analysis, in which case the discretized two-point Pearson correlations computed on the microstructure functions (as given in equation 1) are also needed. These can be computed as
\[ g[n, n' | t] = \frac{1}{|S[t]|} \frac{1}{\sigma[t] \sigma'[t]} \sum_{s \in S} \left( m[n, s] - \mu \right) c[s] \left( m[n', s + t] - \mu' \right) c[s + t] \quad (13) \]

where
\[ \mu = \frac{1}{|S[0]|} \sum_{s \in S} m[n, t] c[s], \quad (14) \]
\[ \mu' = \frac{1}{|S[0]|} \sum_{s \in S} m[n', t] c[s], \quad (15) \]
\[ \sigma[t] = \left\{ \frac{1}{|S[t]|} \sum_{s \in S} \left| m[n, t] - \mu \right|^2 c[s] c[s + t] \right\}^{\frac{1}{2}}, \quad (16) \]
\[ \sigma'[t] = \left\{ \frac{1}{|S[t]|} \sum_{s \in S} \left| m[n', s + t] - \mu' \right|^2 c[s] c[s + t] \right\}^{\frac{1}{2}}. \quad (17) \]

### 3. Preparation of microstructural data

#### 3.1. Data generation by phase field simulation

The microstructure studied is a bicontinuous two-phase mixture with equal volume fractions (50 : 50 mixture) generated by phase field simulations using the conserved Cahn-Hilliard dynamics. Phase field method is a computational technique for simulating phase transformations [14, 15, 16]. It evolves the values of an order parameter \( \phi \) and other spatially varying field quantities of interest by some dynamical equations governing the evolution process. The order parameter has different values in different phases, and smoothly varies across a finite-thickness interface between adjacent phases. The diffuse-interface approach used in the phase field method allows one to apply correct dynamics in different phases without explicitly tracking the locations of the interface. The governing equation that was used to generate the data used in this study is the Cahn-Hilliard equation [11, 17],
\[ \frac{\partial \phi}{\partial t} = L_\phi \nabla^2 \left( \frac{\partial f}{\partial \phi} - \epsilon^2 \nabla^2 \phi \right) \quad (18) \]

where \( L_\phi \) and \( \epsilon \) are the mobility and the gradient energy coefficients, respectively. The bulk free energy, \( f(\phi) \), is assumed to be a double-well potential,
\[ f(\phi) = \frac{W}{4} \phi^2 (1 - \phi)^2 \quad (19) \]
where $W$ is the well-height parameter controlling the height of the double-well potential. The double-well potential given above has two minima at $\phi = 0$ and $\phi = 1$, which represent the equilibrium values of the two phases.

The simulation is performed using a dimensionless form of the Cahn-Hilliard equation, where the variables are scaled by constant scaling factor so that the scaled quantities no longer carry any physical units. Specifically, we define the spatial variable $x = \tilde{x}/l$, where $l$ is a characteristic length, and $t = \tilde{t}/\tau$, where $\tau$ is the associated time scale and the tilde indicates the original dimensional variables. We choose $l$ and $\tau$ such that the grid spacings and the mobility coefficient are one: $\Delta x = \Delta y = \Delta z = 1.0$, and $L_\phi = L_\phi \tau/l^2W = 1.0$. The time step is chosen to be $\Delta t = 0.05$. To generate sufficient statistics for accurate analyses of interfacial morphologies, a large computational domain size of $1024 \times 1024 \times 1024$ on a Cartesian grid was used in the simulation. In the phase field method, the interfacial thickness $\delta$ is determined by the values of $\epsilon^2$ and $W$ in the Cahn-Hilliard equation (equations 18 and 19). In order to ensure numerical stability while maintaining sufficient interfacial resolution, we use $\epsilon^2 = 0.2$ and $W = 0.4$ in the simulation, which gives an interfacial thickness of $\delta = 4.0$. This results in the interfacial region (defined by $\phi$ in the range of 0.1 to 0.9) to be approximately four grid points wide. Random noise of amplitude 0.1 is used as the initial condition for the order parameter $\phi$ in the simulation. The initial input has an average value of $\phi = 0.5$, which results in the 50 : 50 volume fraction formed after spinodal decomposition. Since the Cahn-Hilliard equation is conserved, the volume fractions of the phases remain constant throughout their evolution in the simulation.

The microstructure is evolved for a dimensionless simulation time of $t = 65000$, which is sufficient to produce self-similar structures [18]. The resultant microstructure has a characteristic

**Figure 1:** Three-dimensional plot of the simulated bicontinuous microstructure showing the whole simulation box.
Figure 2: A subregion of the simulated bicontinuous microstructure shown in a cubic domain with side lengths six times the characteristic length scale $S_v^{-1}$. The parameter, $2\Delta$, denotes the spacing between regions of the same phase.

length of $S_v^{-1} = 23.5$, and characteristic time given by $S_v^{-1}/(dS_v^{-1}/dt) = 1.11 \times 10^{-4}$, where $S_v^{-1}$ is the inverse of interfacial area per unit volume. A three-dimensional illustration of the microstructure is given in figure [4] in which the entire periodic simulation box is shown. In this plot, the phase corresponding to order parameter $\phi = 0$ is shown in solid color, whereas the other phase with $\phi = 1$ is shown as transparent. From now on, we will refer to these two phases as the $\alpha$ and $\beta$ phases, respectively. In figure [2], a subregion of the microstructure is shown in a cubic domain. The cubic domain has side lengths equal to six times the characteristic length scale $S_v^{-1}$. From the microstructure plots, we see that the two phases in the microstructure are interconnected throughout the simulation domain, and they also penetrate into each other frequently.

3.2. Calculation of interfacial quantities

In order to capture the interfacial morphology and interfacial dynamics of the microstructure during coarsening, we use the interfacial curvatures and interfacial normal velocity to quantify the microstructure and its evolution. The interfacial curvatures are calculated using a level-set method [19, 20, 21], in which a signed distance function (SDF) is used to implicitly define the interface locations. The SDF gives the distance to the nearest interface at a specific location in space. It has opposite signs on the two sides of the interface, therefore the interface is defined as where the SDF crosses zero. The SDF can be constructed from a binary representation of the two phases, in which the order parameter has only two values – each representing one of the two phases. The construction of the SDF follows an iterative reinitialization routine described in reference [21]. With the SDF, the interfacial mean curvature and Gaussian curvature can be calculated from the spatial derivatives of the SDF [21], and other forms of curvatures can be subsequently calculated from the two. To reduce noise in higher-order derivatives of the SDF, the SDF is smoothed by evolving it in a diffusion process [19]. By comparing the interfacial locations between adjacent time steps, the interfacial normal velocity may be obtained.
In figure 3, the interfacial mean curvature $H$, Gaussian curvature $K$, net curvature $d$, and interfacial normal velocity $V$ are shown for the interface within a subregion of the microstructure. All interfacial quantities are calculated assuming the interfacial normal points from the $\alpha$-phase to the $\beta$-phase, which holds for the rest of this paper.

3.3. Local state selection for two-point statistics calculation

Since the evolution of the interface during coarsening is primarily controlled by the local interfacial mean curvature $H$, we choose the interfacial mean curvature $H$ as the main descriptor for the interfacial morphologies, and the interfacial normal velocity $V$ as the descriptor for interfacial dynamics. In addition, we will use the interfacial net curvature $d$ as an auxiliary to the interfacial mean curvature in describing interfacial morphologies. We will use the interfacial mean curvature $H$, the interfacial net curvature $d$, and the interfacial normal velocity $V$ as the spatially varying interfacial quantities, and build local states from these quantities in the calculation of two-point statistics.

We first use the values of $H$, $d$ and $V$ as local states with continuous range of values, and calculate their spatial correlations on the interface. Then, we discretize these local states into bins with equal amounts of interfacial data points (which approximately results in equal amounts of interfacial area) according to their values, and use these discretized local states to...
build microstructure functions for calculating two-point statistics. Since the two-point Pearson correlation is applicable to both continuous and discrete local states, we will use the two-point Pearson correlation as the form of two-point statistics. We thus use equation (7) to calculate the two-point statistics of continuous local states, and equation (13) for discrete local states. In all of these calculations, periodic boundary conditions are assumed since the simulation is conducted under such boundary conditions. In addition, as these interfacial quantities are only defined on the interface, we use a mask array which allows only statistics on the interface. The mask is constructed such that interfacial voxels have values of 1 and non-interfacial voxels have values of 0.

For comparison, we also use the binary order parameter as the local state in the calculation of two-point statistics. The binary order parameter is defined as

$$\phi_b = \begin{cases} 
0 & \text{if in phase } \alpha, \\
1 & \text{if in phase } \beta.
\end{cases}$$

(20)

The binary order parameter is a representation of the two phases in the microstructure in which the interfaces between phases are infinitely thin. Since it is defined in the bulk phases all over the microstructure, we will use the whole simulation box as the mask in the calculation of the two-point statistics, which effectively means that no mask is applied as the microstructure is periodic.

4. Results

We investigate the bicontinuous microstructure using statistical analysis. First, we examine the statistics of the local interfacial quantities ($H, d,$ and $V$) by considering their univariate distributions. Then, we calculate the spatial distributions of these interfacial quantities using the two-point statistics method.

4.1. Statistics of local interfacial quantities

The probability densities of the three interfacial quantities of interest ($H, d,$ and $V$) are plotted in figure 4. The probability densities are calculated from a triangular mesh along the isosurface where the order parameter crosses zero. Each of the triangular patches on the mesh is associated with a value in one of the interfacial quantities ($H, d,$ and $V$), and the probability densities of the interfacial quantities are weighted by the areas of the patches. Therefore, the probability densities plotted in figure 4 show the interfacial area weighted probability distributions of the interfacial quantities.

From figure 4 we see that the distributions of $H$ and $V$ are perfectly symmetric about zero, which indicates that the microstructure is symmetric with respect to the two phases. This is because the two phases in the microstructure have identical bulk free energies and volume fractions. In other words, if we find an interface with a specific morphology in the microstructure, there should be an equal probability of finding another interface with the same morphology but the two phases on switched sides. On the other hand, the net curvature $d$ only resides on the positive side of the probability distribution due to the way $d$ is defined – it only accounts for the unsigned magnitude of the interfacial curvature.

In order to investigate the relations between the interfacial quantities, we calculate the joint probability distributions of the interfacial quantities. Figure 5a shows the joint probability density of the interfacial principal curvatures $\kappa_1, \kappa_2$. This is the interfacial shape distribution (ISD) commonly used in determining the overall morphologies of the microstructure. On the ISD, the principal curvatures are primarily concentrated in the second quadrant and along the $\kappa_1 = -\kappa_2$ line. This indicates that the interfaces in the microstructure largely constitute of saddle-like shapes with principal curvatures of equal magnitude but opposite signs. In figure 5b
Figure 4: Probability density of (a) interfacial mean curvature $H$, (b) interfacial net curvature $d$, and (c) interfacial velocity $V$. All values are in units of $S_v^{-1}$ (curvature) and $dS_v^{-1}/dt$ (velocity).

the ISD is converted to the joint probability density in the $H - d$ space. In this distribution plot, the probability is clustered along the $H = 0$ line. This means that the distribution in $H$ is much narrower than that in $d$. Moreover, the distribution of $d$ is high at finite values but low at near $d = 0$. This indicates that the majority of the interfaces have near zero mean curvature but finite net curvature. In other words, the microstructure mostly consists of interfaces with saddle shapes rather than flat interfaces.

Next we investigate the local correlation between the interfacial curvatures and the interfacial normal velocity. Figure 5c shows the joint probability density of $H$ and $V$. The plot shows a narrow distribution along a straight line across the second and forth quadrants, which indicates a strong linear relation between $H$ and $V$ which correlates the local mean curvature with the negative of the local interfacial normal velocity. This suggests that the evolution of the interfaces in the bicontinuous microstructure is indeed governed by the Gibbs-Thomson effect. However, the correlation is not perfect as would be expected for Allen-Cahn dynamics [12]. On the other hand, the joint probability density of $d$ and $V$ shown in figure 5d displays a distribution with a much larger spread. This implies a weak correlation between the local values of $d$ and $V$.

4.2. Two-point Statistics of Continuous Interfacial Quantities
After examining the local statistics of the interfacial quantities, we now investigate the spatial distributions of these quantities. We do this through calculating the two-point spatial autocorrelations of the interfacial quantities using the two-point statistics method.

First, we calculate the two-point Pearson auto-correlations of the interfacial mean curvature $H$, interfacial net curvature $d$, and interfacial normal velocity $V$. The results are shown in figures 6b, 6c, and 6d in the form of two-dimensional cross-sections across the origin of the three-dimensional correlation maps. For comparison, we also calculate the auto-correlation of the phases in the microstructure (i.e. the auto-correlation of the binary order parameter $\phi_b$),
which is shown in figure 6a. For clear visualization, the values in these correlation plots are capped at $\pm 0.02$. From figure 6 we can see that the auto-correlation of the phases shows a spherically isotropic correlation pattern within a radius of $5 S_v^{-1}$. Outside of this region, the correlation exhibits random patterns. In a slightly larger range, the auto-correlations of $H$ and $V$ show a similar isotropic correlation pattern, outside of which the correlation appears random as well. As for the auto-correlation of $d$, the isotropic correlation pattern only shows within $r < 3 S_v^{-1}$. Outside of that range, the correlation becomes random. The isotropy of these correlations reflects that the bicontinuous microstructure is isotropic in a statistical sense. This means that the morphological features in the microstructure are randomly oriented all over the microstructure domain.

Due to the isotropy of the correlations, we radially average the two-point Pearson correlations shown in figure 6. This will collapse the three-dimensional correlations into one-dimensional plots. In figure 7a, the radially averaged two-point Pearson auto-correlations of the phases, $H$, $d$ and $V$ are shown. The correlations are truncated at a radius of about $8 S_v^{-1}$. All of the auto-correlations show a large peak with maximum equal to 1 at the origin, which is always the case for Pearson auto-correlations. The correlations then become negative after the first peak, and then they oscillate about zero with diminishing amplitudes as $r$ increases.

In particular, the auto-correlation of the binary order parameter $\phi_b$ shows nearly periodic oscillations about zero with exponentially decaying amplitudes. The positive correlation peaks
**Figure 6:** Two-dimensional cross-sections of the two-point Pearson auto-correlations of (a) binary order parameter (labeled as “Phase1”) (b) interfacial mean curvature $H$ (c) interfacial net curvature $d$, and (d) interfacial normal velocity $V$.

can be explained as the correlations between different portions of the same phase, and the negative local minima are results from the anti-correlations between opposite phases. One representation of the structure is that it is, on average, a series of channels where the distance between neighboring channels of the same phase is given by the location of the first local maximum, i.e. $r = 2.8 S_v^{-1}$. Following this logic, the distance between neighboring channels of opposite phases, or the inter-phase distance $\Delta$, is given by half of the first local maximum location – i.e. $\Delta = 1.4 S_v^{-1}$. On the other hand, the first local minimum appears at $r = 1.6 S_v^{-1}$, which is smaller than the inter-phase distance $\Delta$ obtained above. This is because the wide self-correlation peak at $r = 0$ shifts the first local minimum to the right. Since the three-dimensional microstructure consists of channel structures randomly oriented in space, the spatial correlation along the cross-axial direction of one channel may be along the axial direction of some other channel with a different orientation. Therefore, the primary correlation peak also includes the self-correlation along the axial direction of the channels, which widens the peak.

At larger $r$, the spacing between the local minima and maxima become smaller, which seems to be contradictory to the periodicity of the structure. This again can be explained by the three-dimensionality of the microstructure. In two or more dimensions, the distance between second nearest neighbors may be smaller than twice of the first nearest neighbor distance, because the second and first nearest neighbors and the reference point may not be on a straight line. For
**Figure 7:** Radially averaged two-point Pearson correlations of binary order parameter, interfacial curvatures, and interfacial normal velocity. (a) Auto-correlations of the binary order parameter (labeled as “Phase1”), interfacial mean curvature $H$, interfacial net curvature $d$, and interfacial normal velocity $V$. (b) Correlations of the interfacial mean curvature $H$ vs. the negative of interfacial normal velocity $-V$ (blue), and the interfacial net curvature $d$ vs. the absolute value of interfacial normal velocity $|V|$ (red). (c-d) Zoomed-in views of (a-b).

Example, in a three-dimensional cubic lattice with lattice constant $a$, the distance between first nearest neighbors is $a$, second nearest neighbors $\sqrt{2}a$, and third nearest neighbors $\sqrt{3}a$.

In contrast to the binary order parameter, the spatial auto-correlation of the interfacial mean curvature $H$ shows a less periodic pattern. In particular, the auto-correlation of $H$ shows a wide region of anti-correlation from $2.2 S_v^{-1}$ to $3.5 S_v^{-1}$. At $r > 3.5 S_v^{-1}$, the auto-correlation of $H$ is perfectly out of phase with the auto-correlation of $\phi_b$. In other words, the two correlations have minima and maxima at the same locations but with opposite signs, and they also share the same nodes. The first local maximum appears at $r = 4 S_v^{-1}$. From the auto-correlation of $\phi_b$ we know that this distance is about three times the inter-phase distance, or $3\Delta$. Since the interfacial mean curvature is only defined on the interface, the two-point spatial auto-correlation of $H$ will only include interfacial locations. Therefore, in this context, the distance of $3\Delta$ would be the distance between the interfaces on the opposite sides of two neighboring channels of the same phase and the auto-correlation pattern of $H$ could come from a microstructure with two channels of the same phase bending towards opposite directions. In this case, the high positive curvatures on the outer interfaces will correlate with each other, resulting in the peak at $r = 3\Delta$; whereas the
negative curvatures on the inner interfaces will anti-correlate with the outer interfaces, resulting in the two negative local minima at around \( r = \Delta \) and \( r = 2\Delta \). Again the first local minimum is right-shifted due to the wide primary peak caused by the self-correlation effect. After the first positive local maximum, the auto-correlation of \( H \) oscillates with the periodicity of the microstructure, and quickly decays to zero.

The auto-correlation of the interfacial normal velocity \( V \) exhibits the same overall trend as that of \( H \). Specifically, it also has negative local minima at about \( r = \Delta \) and \( r = 2\Delta \), and positive local maximum at around \( r = 3\Delta \), with smaller oscillatory peaks to follow. The similarity between the auto-correlations of \( H \) and \( V \) directly follows from the fact that \( H \) and \( V \) are strongly (anti-)correlated locally. Nevertheless, the auto-correlation of \( V \) also shows some distinct features. In particular, the first local minimum is larger in magnitude and appears at a shorter \( r \), and the local maximum at around \( \sim 2.2 \) \( S^{-1}_v \) becomes positive. This indicates that the relative spatial variations of \( V \) is larger than that of \( H \), and \( V \) changes sign more frequently than \( H \). The positive correlation at \( \sim 2.2 \) \( S^{-1}_v \) implies that the local correlation between \( V \) and \( H \) may be reversed at certain locations on the interface. This is possible, because the
competition among neighboring interfaces during interfacial evolution may cause the movement of the interfaces near an interface with a large curvature to be suppressed or even reversed.

Different from all of the three auto-correlations presented above, the auto-correlation of $d$ shows an oscillatory pattern with a period about half of the other three. This is because the interfacial net curvature $d$ is an unsigned value, and hence interfaces with high positive and high negative mean curvatures will positively correlate with each other in $d$ even though they have opposite signs in $H$ (and also $V$). In fact, the first correlation peak after then primary peak appears at $r = 1.4 S_v^{-1}$, which is exactly the inter-phase distance $\Delta$. This implies that this positive correlation comes from the correlation between the two walls of the channel, which both have high curvatures but with opposite signs. In addition, the auto-correlation of $d$ decays quickly to zero after $r = 4 S_v^{-1}$, which indicates that the interfacial net curvature has a much shorter spatial correlation compared to the phases, the interfacial mean curvature and the interfacial normal velocity.

4.3. Two-point statistics of discretized interfacial quantities

While the two-point statistics presented in the previous section show spatial correlations of the values of the interfacial quantities in general, they do not provide information on the contributions of the subranges in these values to the correlations. Specifically, the two-point statistics of the continuous interfacial quantities does not tell us how the interfaces with high, intermediate, and near zero values of these quantities correlate in space. We investigate such correlations in the current section.

We select 10% bins from the probability distributions of the interfacial curvatures and normal velocity, each of which contain 10% of the total interface, but with different values of the interfacial quantities. Since the two phases in the bicontinuous microstructure are equivalent, the two sides of the interface are completely symmetric, in terms of both morphology and dynamics. Therefore, we can examine only the interfaces with positive interfacial mean curvature $H$ and negative interfacial normal velocity $V$, and the results should also apply to the other half of the interface. Nevertheless, the interfacial net curvature $d$ is defined in an unsigned form. In order to distinguish between positive and negative curvatures while using $d$, we multiply the sign of the interfacial mean curvature $H$ to the value of $d$, which gives the signed interfacial net curvature $\text{sign}(H)d$.

Based on these definitions, five 10% bins from the probability distributions of $H$, $-V$ and $\text{sign}(H)d$ are selected. As illustrated in figure 8, these bins are the 45%–55% (blue), 60%–70% (light blue), 70%–80% (green), 80%–90% (yellow), and 90%–100% (red) percentiles of $H$, $V$, and $\text{sign}(H)d$, which represent the 10% interfaces with near zero (blue), high positive (red), and intermediate positive (light blue, green, yellow) values in the three interfacial quantities. We then construct discretized local states using these 10% bins of the interfacial quantities, and build binary microstructure functions as spatial indicators of the discrete local states, as described earlier. These binary microstructure functions are used in the calculation of the two-point statistics of the 10% bins, in which equation 15 is used.

Figure 9 shows the radially averaged results of the two-point Pearson auto-correlations of the five 10% bins in $H$, $-V$ and $\text{sign}(H)d$. In this figure, the auto-correlations of $H$ and $-V$ both show that the auto-correlations of the largest values of $H$ and $V$ are the most spatially correlated among all values of $H$ and $V$. Moreover, the spatial auto-correlations of the largest values of $H$ and $-V$ show approximately the same correlation patterns as the correlation of $H$ and $V$ when continuous values of them are used. For the near zero bins of $H$ and $-V$, the auto-correlation decays monotonously to zero within $r \leq 2 S_v^{-1}$. The auto-correlations of the other bins with intermediate values of $H$ and $-V$ follow the same trend as the largest bins. Specifically, they have local minima and maxima at the same locations but with decreasing amplitudes as we go to smaller value bins, and they also share the same nodes. This indicates that most of the
**Figure 9:** Radially averaged two-point Pearson auto-correlations of interfaces with values of (a,b) interfacial mean curvature $H$, (c,d) signed interfacial net curvature $\text{sign}(H)d$, and (e,f) negative interfacial normal velocity $-V$, in the 45% – 55% (blue), 60% – 70% (light blue), 70% – 80% (green), 80% – 90% (yellow), and 90% – 100% (red) percentiles.
spatial correlation in $H$ and $V$ come from the extreme values. Interfaces with large (positive or negative) values of $H$ and $V$ form clusters in the microstructure. The average distance between these clusters is given by the first significant local maximum on the correlation curve, i.e. $4 S_v^{-1}$, or $\sim 3\Delta$. In between these interfaces with large $H$ and $V$, the interface smoothly varies in $H$ and $V$, which results in a similar correlation pattern of the intermediate values in $H$ and $V$. For the interfaces with near zero values of $H$ and $V$, it shows that they are randomly distributed in space.

In contrast to the auto-correlations of the bins in $H$ and $-V$, the auto-correlations of the five bins in $\text{sign}(H)d$ do not share the same patterns. In particular, the high positive values of $\text{sign}(H)d$ show a similar auto-correlation pattern as high positive $H$. This is because all interfaces with large $H$ will also have large $d$, although the inverse statement is not necessarily true. However, as is shown in figure 9, the interfaces with large $d$ but smaller or near zero $H$ are either correlated in the same way as the largest $H$, or not correlated in the long range at all. Therefore, the net result is that the auto-correlation of large $\text{sign}(H)d$ shows a similar correlation pattern as that of large $H$. For the near zero bin of $\text{sign}(H)d$, a positive correlation peak appears at about $r = 1.4 S_v^{-1}$, or $1\Delta$, on the auto-correlation curve. This indicates that interfaces within this bin, i.e. flat interfaces, are clustered in the microstructure with an average inter-cluster distance equal to the average inter-phase distance. Unlike the corresponding bins in $H$ and $-V$, the bins of $\text{sign}(H)d$ with intermediate values do not have the same local minima, local maxima, or nodes. In addition, in the range of $r = 2.2 - 3 S_v^{-1}$, the auto-correlation of the intermediate bins of $\text{sign}(H)d$ show negative correlations, while the near zero and high positive bins are not correlated in this range.

5. Conclusions

The spatial distribution of the interfacial quantities and the bulk phases is determined using two-point statistics. From the spatial correlation of the order parameter or bulk phases, we find that the average distance between neighboring regions in opposite phases is $\Delta = 1.4 S_v^{-1}$. Using two-point statistics of interfacial quantities, we demonstrate that the relative spatial variations of $V$ are larger than those of $H$, and $V$ changes signs more frequently than $H$ with distance from a point in space. This large spatial variation in interfacial velocity is likely caused by the excess or depletion of solute induced by the large velocity of interfaces with high curvatures. This also leads to large velocity of flat interfaces nearby. In addition, for interfaces with high positive and high negative values of $H$, the signed total curvature $\text{sign}(H)d$ and $V$ tend to be spatially localized. The average spacing between regions with the same sign of curvature or velocity is $\sim 4 S_v^{-1}$ or $\sim 3\Delta$. Interfaces with near zero values of $H$ are only correlated over very short distances interfaces with near zero $d$ are clustered in space, with an average inter-cluster spacing of $1.4 S_v^{-1}$, or $1\Delta$. The spatial distribution of interfaces with near zero $V$ largely resembles that of near zero $H$ interfaces.

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