

Higher-order orientational effects on geometric percolation in two-dimensional stick networks

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We investigate geometric percolation in two-dimensional assemblies of randomly positioned, widthless sticks of uniform length. The stick orientations are sampled from probability density functions with a vanishing mean orientational order parameter, while their higher-order structure varies continuously from perfect isotropy to perfect cross-alignment. Monte Carlo simulations show that the percolation threshold is lowest for the isotropic distribution and increases toward the perfectly cross-aligned limit. This variation is strongly nonuniform: small orientational perturbations near perfect cross-alignment produce substantially larger changes in the threshold than comparable perturbations near isotropy. The simulation results are interpreted using an orientation-resolved eigenvalue formulation based on the full excluded-area kernel and a fully preaveraged excluded-area approximation. For the probability density functions considered, the two formulations predict nearly identical thresholds. Orientation-dependent variations in the expected contact numbers therefore provide only a minor correction, whereas the dominant orientational dependence is retained by the averaged excluded area between pairs of sticks. A harmonic representation of this quantity further shows that the second moment captures the leading variation near isotropy, while progressively higher-order moments are required as the distribution approaches perfect cross-alignment. The results identify the orientational information that must be retained to predict geometric percolation in distributions having the same mean order parameter but different higher-order structures.

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I. INTRODUCTION

Percolation controls the emergence of long-range connectivity and transport in conductor–insulator composites, with high-aspect-ratio conductive inclusions such as fibers, nanowires, and nanotubes being particularly effective in forming connected networks at low filler contents [1–3]. Conducting nanowire (NW) networks are a representative example, and their use in flexible and transparent electronics requires high electrical conductivity without excessive loss of optical transparency or mechanical compliance [4–6].

The response of an NW assembly depends not only on the number of nanowires, but also on how they are distributed in space and orientation. Orientational order affects both the percolation threshold and the directional character

of the resulting network [7–9]. This has motivated growing interest in nanostructured technologies based on cross-aligned NW architectures, for which transport and functional properties are strongly influenced by network connectivity. Such architectures have been reported to provide better electrical and optical performance than randomly oriented networks [10–14]. From the standpoint of geometric percolation alone, however, this trend can appear counterintuitive, since uniformly distributed orientations are expected to yield a lower percolation threshold. This apparent contrast arises because device performance depends on more than geometric connectivity. Junction resistance and other transport parameters [15,16], as well as the spatial uniformity and network structure generated during deposition [15], also influence the response of a real NW network. The model considered below isolates the contribution of orientational order to geometric connectivity and does not include these additional effects.

We consider two-dimensional assemblies of randomly positioned, widthless sticks of uniform length that are allowed to intersect. Their orientations are sampled from probability density functions (PDFs) with a vanishing mean orientational order parameter. The distributions span the range from perfect isotropy, represented by a uniform angular distribution, to perfect cross-alignment, in which the sticks are equally divided between two orthogonal directions. The two limiting cases therefore have the same mean orientational order parameter but markedly different higher-order orientational

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structures. They provide a controlled setting for determining which descriptors of orientational order govern the percolation threshold.

The analysis addresses two related questions. The first is whether the orientational dependence of the threshold is determined primarily by the excluded area averaged over mutual stick orientations. The second is whether orientation-dependent variations in the expected contact numbers provide a significant additional contribution. Monte Carlo simulations are compared with two theoretical descriptions: an orientation-resolved eigenvalue formulation based on the full excluded-area kernel and a fully preaveraged excluded-area approximation. A harmonic representation of the averaged excluded area is subsequently used to express its dependence on successive moments of the orientational order parameter.

The simulation results show that the percolation threshold is lowest for the isotropic distribution and increases toward the perfectly cross-aligned limit, consistent with previous studies [15,17,18]. The threshold is considerably more sensitive to orientational perturbations close to perfect cross-alignment than close to isotropy. For the PDFs examined here, the orientation-resolved and fully preaveraged formulations produce nearly identical predictions. Orientation-dependent variations in the expected contact numbers therefore provide only a small correction, while the dominant orientational dependence is retained by the averaged excluded area.

Near isotropy, the leading variation is adequately described by the second moment of the orientational order parameter. Progressively higher-order moments are required as the distribution approaches perfect cross-alignment. The remaining difference between the theoretical and simulated absolute threshold values is consistent with the role of closed loops in two-dimensional stick networks. These results identify the orientational information that must be retained to predict geometric percolation across distributions that share the same mean order parameter but differ in their higher-order structure.

II. MODELS AND METHODS

The orientation-resolved formulation, the fully preaveraged approximation, and the Monte Carlo simulations considered below refer to the same physical model: a two-dimensional assembly of penetrable, widthless sticks of identical length L , whose centers and orientations are statistically independent. In the numerical simulations, N such sticks are placed in a square domain of side Λ , with their centers sampled independently from a uniform spatial distribution. The number density is $\rho = N/\Lambda^2$, and the corresponding dimensionless density is $n = \rho L^2$. Throughout this work, we report the percolation threshold in dimensionless form as $n_c = \rho_c L^2$.

Let $\theta \in [-\pi/2, \pi/2)$ denote the unoriented angle of a stick measured from a fixed in-plane axis, and define $S = \cos(2\theta)$. To isolate the role of orientational fluctuations while maintaining a vanishing mean orientational order parameter, we focus on ensembles satisfying

$$\langle S \rangle = \langle \cos(2\theta) \rangle = 0, \quad (1)$$

where $\langle \cdot \rangle$ denotes the average with respect to the orientation distribution. We next introduce three distinct PDFs that satisfy this condition.

A. Probability density functions for stick orientations

We consider three orientation distributions: Maltese-cross, power, and exponential distributions. Each distribution is associated with a family of PDFs, denoted $\psi(\theta, \kappa)$, whose parameter κ allows the family to span the range from perfect isotropy, corresponding to a uniform angular distribution, to the perfect-cross distribution, in which half of the sticks are oriented along a given direction and the other half along the orthogonal direction. For each admissible value of the parameter κ , a normalization factor ensures that

$$\int_{-\pi/2}^{\pi/2} \psi(\theta, \kappa) d\theta = 1. \quad (2)$$

The second moment $\langle S^2 \rangle = \langle \cos^2(2\theta) \rangle$ is equal to $\langle S^2 \rangle_{\text{iso}} = 1/2$ and $\langle S^2 \rangle_{\text{pc}} = 1$ in the isotropic and perfect-cross limits, respectively. The subscript iso denotes the uniform PDF $\psi_{\text{iso}}(\theta) = 1/\pi$, while pc denotes the perfect-cross limit. The three PDF families vary in their higher-order orientational structure while maintaining $\langle S \rangle = 0$. With the π -periodic extension implied by the unoriented angle, they also satisfy

$$\psi(\theta, \kappa) = \psi(-\theta, \kappa), \quad \psi(\theta, \kappa) = \psi\left(\frac{\pi}{2} - \theta, \kappa\right). \quad (3)$$

It therefore suffices to specify each PDF over $0 \leq \theta \leq \pi/4$.

a. Maltese-cross distribution. The associated PDF family is given by

$$\psi_{\text{mc}}(\theta, \alpha) = \begin{cases} \frac{1}{4\alpha}, & 0 \leq \theta \leq \alpha, \\ 0, & \alpha < \theta \leq \frac{\pi}{4}. \end{cases} \quad (4)$$

Equation (4) applies for $0 < \alpha \leq \pi/4$. The perfectly cross-aligned distribution is approached as $\alpha \rightarrow 0$, whereas the isotropic distribution is obtained at $\alpha = \pi/4$. The second moment of the orientational order parameter for this PDF is

$$\langle S^2 \rangle = \langle \cos^2(2\theta) \rangle = \frac{1}{2} \left(1 + \frac{\sin(4\alpha)}{4\alpha} \right).$$

b. Power distribution. The corresponding PDF family is given by

$$\psi_{\text{pow}}(\theta, m) = A_{\text{pow}}(m) |\cos(2\theta)|^m, \quad (5)$$

where $A_{\text{pow}}(m)$ is the normalization factor

$$A_{\text{pow}}(m) = \frac{\Gamma(m+1)}{2^m [\Gamma(\frac{m+1}{2})]^2}.$$

The exponent satisfies $0 \leq m < \infty$. The isotropic distribution is obtained at $m = 0$, whereas perfect cross-alignment is approached as $m \rightarrow \infty$. The second moment of the orientational order parameter for this PDF is

$$\langle S^2 \rangle = \langle \cos^2(2\theta) \rangle = \frac{m+1}{m+2}.$$

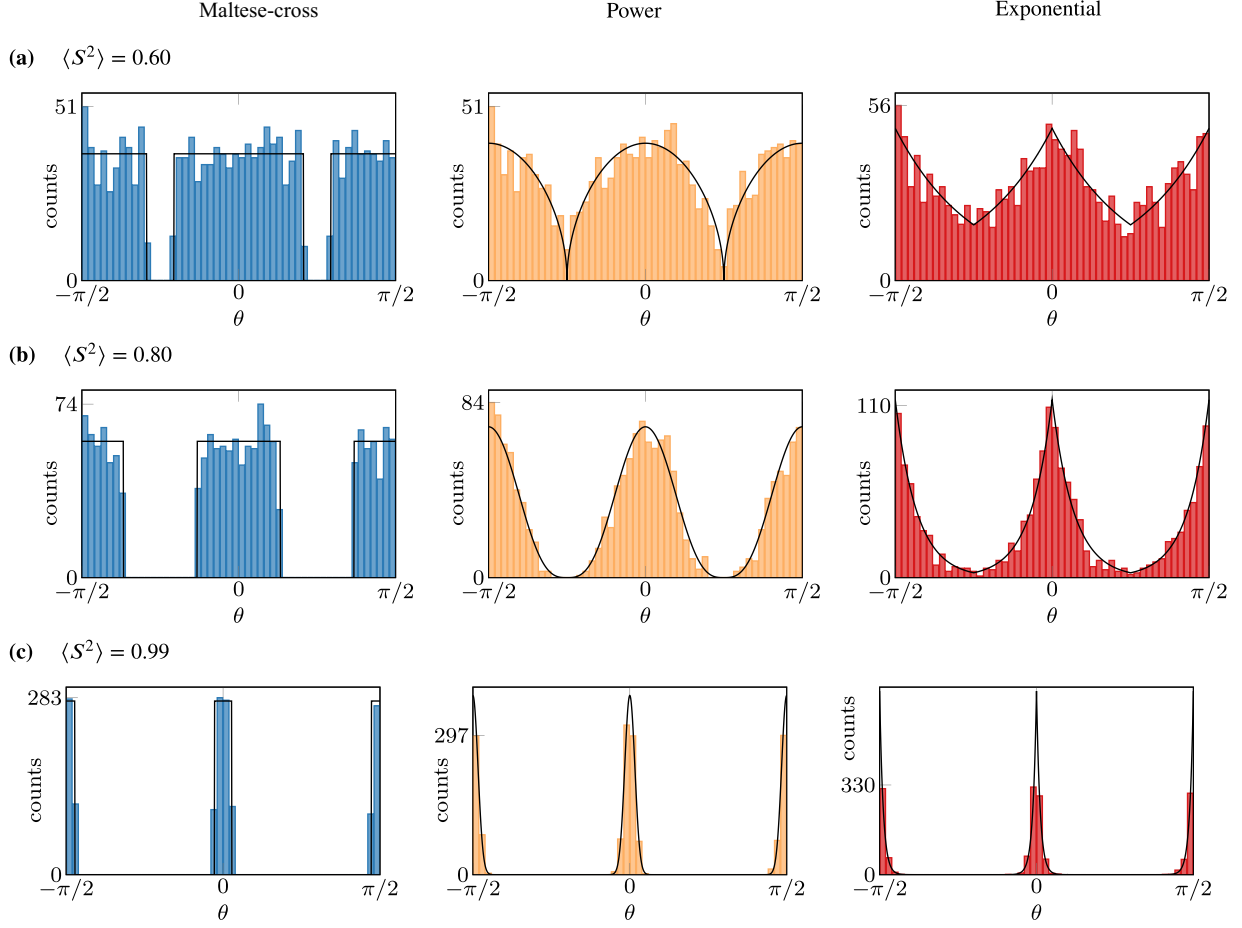


FIG. 1. Examples of discrete stick-orientation distributions used in the numerical simulations (Sec. II C). Orientations $\theta \in [-\pi/2, \pi/2]$ are sampled from the Maltese-cross, power, and exponential PDFs for (a) $\langle S^2 \rangle = 0.6$, (b) $\langle S^2 \rangle = 0.8$, and (c) $\langle S^2 \rangle = 0.99$. Each histogram contains approximately 1500 sampled orientations, corresponding to the number of sticks N at $n \approx 6$ in a domain of size $\Lambda = 16L$, and is divided into 50 bins. The black curves show the continuous PDFs defined by Eqs. (4)–(6). The parameters are selected to match the prescribed value of $\langle S^2 \rangle$: $\alpha = \{6.49 \times 10^{-1}, 4.15 \times 10^{-1}, 8.69 \times 10^{-2}\}$, $m = \{5.00 \times 10^{-1}, 3.00, 9.80 \times 10^1\}$, and $\beta = \{1.28, 4.59, 2.80 \times 10^1\}$. To express the continuous PDFs on the histogram count scale, they are multiplied by $N\pi/50$, where $\pi/50$ is the histogram-bin width.

c. Exponential distribution. The corresponding PDF family is given by

$$\psi_{\text{exp}}(\theta, \beta) = A_{\text{exp}}(\beta) e^{-\beta\theta}, \quad 0 \leq \theta \leq \frac{\pi}{4}, \quad (6)$$

where $A_{\text{exp}}(\beta)$ is the normalization factor

$$A_{\text{exp}}(\beta) = \frac{\beta}{4(1 - e^{-\beta\pi/4})}.$$

Equation (6) applies for $\beta > 0$. The isotropic PDF is recovered by continuity as $\beta \rightarrow 0$, for which $A_{\text{exp}} \rightarrow 1/\pi$, whereas perfect cross-alignment is approached as $\beta \rightarrow \infty$. The second moment of the orientational order parameter for this PDF is

$$\langle S^2 \rangle = \langle \cos^2(2\theta) \rangle = \frac{1}{2} \left[1 + \frac{\beta^2(1 + e^{-\pi\beta/4})}{(\beta^2 + 16)(1 - e^{-\pi\beta/4})} \right].$$

Higher moments of the order parameter, such as $\langle S^4 \rangle$ and $\langle S^6 \rangle$, can be obtained for these three PDFs in closed form. Because the three PDF families are controlled by different parameters, we use $\langle S^2 \rangle$ as a common measure of the departure from isotropy. Within each family, $\langle S^2 \rangle$ varies monotonically between 1/2 in the isotropic limit and 1 in the perfect-cross

limit, and therefore uniquely identifies a member of that family. The same value of $\langle S^2 \rangle$, however, does not in general identify the same orientational distribution across different families. Examples of realizations of the PDFs in Eqs. (4)–(6) for $\langle S^2 \rangle = 0.6, 0.8$, and 0.99 are presented in Fig. 1. Figure 2 shows schematic illustrations of arrangements of sticks whose

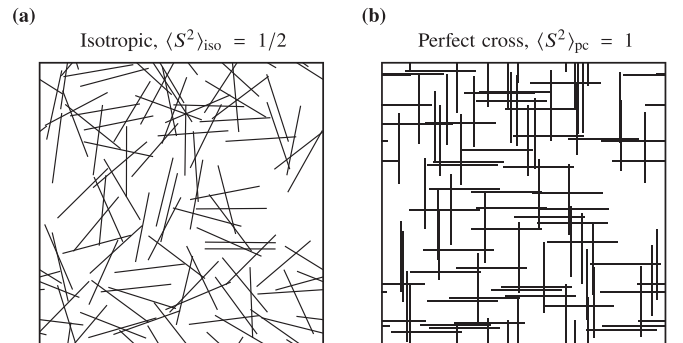


FIG. 2. Schematic stick arrangements sampled from (a) the isotropic PDF and (b) the perfect-cross PDF.

orientations are drawn from the isotropic and perfect-cross PDFs.

B. Orientation-resolved and preaveraged excluded-area descriptions of the percolation threshold

To separate the effect of orientation-dependent variations in the expected contact numbers from that of the averaged contact geometry, we compare two related theoretical descriptions: an orientation-resolved eigenvalue formulation based on the full excluded-area kernel, and a fully preaveraged excluded-area approximation. In the following theoretical developments, a specific member of any of the PDF families introduced above is considered fixed. We therefore write the orientational distribution as $\psi(\theta)$, suppressing its dependence on the corresponding family parameter.

1. Orientation-resolved eigenvalue formulation

The orientation-resolved eigenvalue formulation follows the general framework developed recently for anisotropic penetrable particles [19], which builds on the random-geometric-graph approach introduced in Ref. [20] for isotropically oriented slender rods in three dimensions. Here, the formulation is specialized to two-dimensional widthless sticks and to the orientational distributions described in Sec. II A. It retains the dependence of the expected contact numbers on the pair of orientations θ_i and θ_j , rather than replacing it with a single preaveraged value, while assuming a locally tree-like network and therefore neglecting closed loops.

The resulting formulation is semianalytical: discretization of the angular variable reduces the orientation-dependent connectivity condition to a finite-dimensional matrix eigenvalue problem, whose largest eigenvalue is then evaluated numerically. The equations below recast the random-geometric-graph formulation of Ref. [20] as an eigenvalue problem and present its specialization to the present two-dimensional setting and to PDFs that are not necessarily isotropic.

To construct the finite-dimensional eigenvalue problem, we divide the angular interval $[-\pi/2, \pi/2)$ into N_θ bins of uniform width $\Delta\theta = \pi/N_\theta$. We use the bin midpoints $\theta_i = -\pi/2 + (i + 1/2)\Delta\theta$ with $i = 0, \dots, N_\theta - 1$ as the representative orientations. The probability mass assigned to angular bin i is approximated by midpoint quadrature and explicitly renormalized so that $\sum_i p_i = 1$. Since all bins have the same width, the normalized discrete weights can be written as

$$p_i = \frac{\psi(\theta_i)\Delta\theta}{\sum_\ell \psi(\theta_\ell)\Delta\theta} = \frac{\psi(\theta_i)}{\sum_\ell \psi(\theta_\ell)}, \quad \sum_i p_i = 1.$$

The expected number of sticks in angular bin j that overlap a reference stick in bin i is denoted by z_{ij} and is given by

$$z_{ij} = \rho p_j a_{ij}, \quad (7)$$

where a_{ij} denotes the excluded area between a pair of sticks with orientations θ_i and θ_j . For widthless sticks of uniform length L , the excluded area a_{ij} satisfies

$$a_{ij} = L^2 |\sin(\gamma_{ij})|.$$

The dependence of a_{ij} on the relative angle $\gamma_{ij} = \theta_j - \theta_i$ between sticks i and j generates the orientation-resolved

variation in the expected contact numbers through Eq. (7). Retaining this dependence avoids replacing the contact numbers by a single value preaveraged over the orientational PDF.

For tree-like network structures, the dimensionless percolation threshold is the smallest value of n for which the solution vector $\{\epsilon_k\}$ of the system of linear equations

$$\epsilon_j = \rho \sum_k a_{jk} p_k \epsilon_k = n \sum_k p_k |\sin(\gamma_{jk})| \epsilon_k \quad (8)$$

is nonzero [20]. The quantity ϵ_k in Eq. (8) is the probability that a randomly selected stick in angular bin k belongs to the incipient percolating cluster, or giant component, as n approaches the percolation threshold from above; this definition corresponds to Eq. (10) of Ref. [20]. An equivalent condition can be expressed in terms of eigenvalues by defining (i) the new, rescaled variables $\eta_j = \sqrt{p_j} \epsilon_j$, and (ii) the real symmetric matrix $A = [A_{jk}]$, whose non-negative elements are

$$A_{jk} = \sqrt{p_j p_k} |\sin(\gamma_{jk})|.$$

In terms of these variables, the condition in Eq. (8) can be restated as the problem of finding the smallest value of n for which the equations

$$\sum_k A_{jk} \eta_k = \frac{1}{n_c} \eta_j \quad (9)$$

have nonzero solutions for the $\{\eta_j\}$. Equation (9) shows that finding the percolation threshold is equivalent to evaluating the largest eigenvalue $e_{\max}$ of the matrix A , that is,

$$n_c = \frac{1}{e_{\max}}. \quad (10)$$

The largest eigenvalue of the resulting matrix A was computed using the iterative power method [21]. All eigenvalue results reported below were obtained with $N_\theta = 4096$ angular bins. Repeating the calculations with $N_\theta = 2048$ produced no discernible differences in the reported results.

2. Fully preaveraged excluded-area approximation

In the fully preaveraged approximation, the dependence of the contact kernel on the individual stick orientations is removed before solving the connectivity problem. The orientation-resolved factor $|\sin(\gamma_{ij})|$ is replaced by its average $\langle |\sin(\gamma_{ij})| \rangle$ over the prescribed orientational PDF. This orientationally averaged excluded-area factor is

$$\langle |\sin(\gamma_{ij})| \rangle = \int_{-\pi/2}^{\pi/2} \int_{-\pi/2}^{\pi/2} \psi(\theta_i) \psi(\theta_j) |\sin(\gamma_{ij})| d\theta_j d\theta_i. \quad (11)$$

Replacing the orientation-resolved factor $|\sin(\gamma_{jk})|$ in Eq. (8) by this scalar quantity gives

$$n_c = \frac{1}{\langle |\sin(\gamma_{ij})| \rangle}. \quad (12)$$

Equation (12) has been obtained previously from excluded-volume arguments [1] and a Bethe-lattice approach [18], both of which neglect the orientation dependence of the expected contact numbers. The effective medium approximation (EMA) derived in the Appendix retains the same inverse

dependence on $\langle |\sin(\gamma_{ij})| \rangle$, but yields an overall coefficient twice as large. The corresponding averaged excluded area is

$$\langle a \rangle = L^2 \langle |\sin(\gamma_{ij})| \rangle. \quad (13)$$

The orientationally averaged excluded-area factor $\langle |\sin(\gamma_{ij})| \rangle$ from Eq. (11) takes the values $2/\pi$ and $1/2$ in the isotropic and perfectly cross-aligned limits, respectively.

C. Monte Carlo simulations of the percolation threshold

We perform Monte Carlo simulations of two-dimensional stick networks to assess the dependence of the percolation threshold on $\langle S^2 \rangle$ for the orientational PDFs introduced in Sec. II A. A summary of the procedure used to generate the numerical samples and determine the percolation threshold is given below; further computational details can be found in Ref. [22].

a. Geometry generation. Each numerical realization is generated according to the stick model defined at the beginning of this section. The acceptance–rejection method is used to sample each stick orientation from one of the PDFs presented in Sec. II A. Figure 1 shows sampled-orientation histograms at $\langle S^2 \rangle = 0.6, 0.8$, and 0.99 .

Periodic images of the simulation cell are used only when evaluating stick intersections near the domain boundaries, thereby avoiding an artificial reduction in the local stick density close to the edges; percolation is assessed from the connectivity of the resulting stick-intersection graph.

b. Percolation threshold. We identify the percolation threshold using a percolation cluster search [15,17,23,24]. For each numerical realization, a graph is constructed in which the sticks are the nodes and an edge connects each pair of intersecting sticks. A realization is classified as percolating when a connected component of this graph contains sticks intersecting two opposite boundaries of the simulation domain. Percolation then occurs in the direction perpendicular to those boundaries. We generate 500 numerical realizations for each value of the dimensionless density n and compute the percolation probability P as the fraction of percolating realizations. We consider $4 \leq n \leq 8$; for every combination of the PDF family and $\langle S^2 \rangle$, the percolation probability changes from 0 to 1 within this interval. The simulated percolation probabilities are fitted with the logistic function

$$P(n) = [1 + \exp(c_1(c_2 - n))]^{-1},$$

where c_1 and c_2 are fitting parameters. Since $P(c_2) = 0.5$, the corresponding finite-domain estimate is $n_{P=0.5} = c_2$. The procedure is repeated for $\Lambda = 4L, 8L$, and $16L$, and the threshold n_c in the thermodynamic limit is estimated by fitting the finite-domain results to

$$n_{P=0.5}(\Lambda) - n_c \propto \left(\frac{\Lambda}{L}\right)^{-1.6}. \quad (14)$$

We explore the range of $\langle S^2 \rangle$ values from $\langle S^2 \rangle_{\text{iso}} = 1/2$ to $\langle S^2 \rangle_{\text{pc}} = 1$ using the Maltese-cross, power, and exponential PDFs. We observe that the estimated percolation threshold depends only weakly on the exponent in Eq. (14): varying the exponent between -2.50 and -0.75 produces only minor changes over the entire range of $\langle S^2 \rangle$ and for all PDFs considered. The value -1.6 in Eq. (14), which provides the

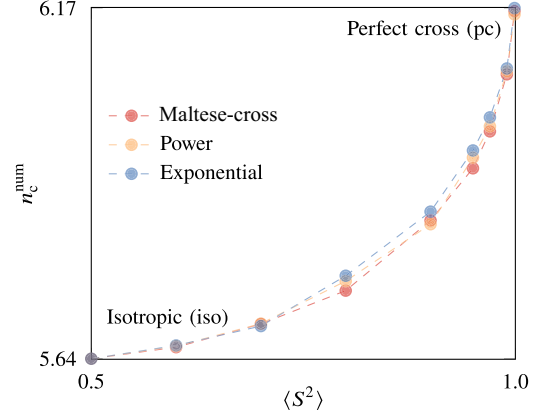


FIG. 3. The dimensionless percolation threshold n_c^{num} obtained from Monte Carlo simulations is shown as a function of $\langle S^2 \rangle$ for the Maltese-cross, power, and exponential PDFs introduced in Sec. II A.

best fit to all datasets, is consistent with values reported in the literature [25,26].

III. RESULTS

In this section, we compare the percolation thresholds obtained from Monte Carlo simulations with the predictions of the orientation-resolved eigenvalue formulation and the fully preaveraged approximation. We first examine the threshold variation across the three PDF families, then quantify the effect of retaining the orientation dependence of the expected contact numbers, and finally interpret the dominant orientational dependence in terms of successive angular harmonics and moments.

We use the superscripts eig, pre, and num for quantities obtained from the eigenvalue formulation, Eq. (10), the fully preaveraged approximation, Eq. (12), and the Monte Carlo simulations described in Sec. II C, respectively. With abuse of notation, we indicate the percolation threshold in the isotropic ($\langle S^2 \rangle = \langle S^2 \rangle_{\text{iso}}$) and perfect-cross ($\langle S^2 \rangle = \langle S^2 \rangle_{\text{pc}}$) limits with n_{iso} and n_{pc} , respectively. For a generic value of $\langle S^2 \rangle$, the percolation threshold is denoted by n_c .

To facilitate the comparison of the results, we introduce two rescalings of the percolation threshold. For each evaluation approach $\mu \in \{\text{eig, pre, num}\}$, we define

$$\tilde{Y}^\mu = \frac{n_c^\mu - n_{\text{iso}}^\mu}{n_{\text{pc}}^\mu - n_{\text{iso}}^\mu}, \quad (15)$$

and

$$Y^\mu = \tilde{Y}^\mu \frac{n_{\text{pc}}^\mu}{n_c^\mu}. \quad (16)$$

Both rescalings map the threshold values onto the interval $[0,1]$, with the isotropic and perfectly cross-aligned limits corresponding to 0 and 1, respectively.

A. Simulated thresholds and comparison with theory

Figure 3 shows the percolation threshold obtained for the PDFs introduced in Sec. II A using the Monte Carlo simulations described in Sec. II C. For all three PDFs, the percolation threshold increases monotonically from

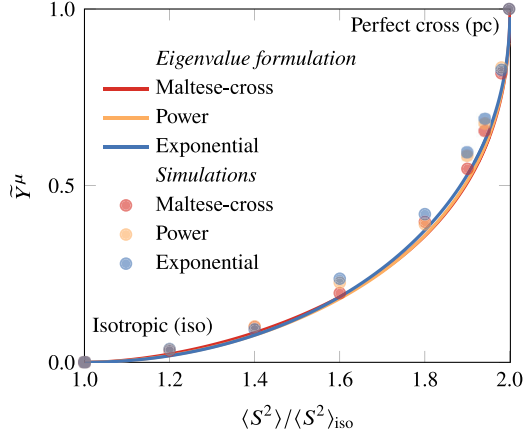


FIG. 4. The rescaled threshold $\tilde{\gamma}^\mu$ defined in Eq. (15) is shown as a function of $\langle S^2 \rangle / \langle S^2 \rangle_{\text{iso}}$ for the Maltese-cross, power, and exponential PDFs. Here, $\mu = \text{num}$ for the Monte Carlo simulations (symbols) and $\mu = \text{eig}$ for the eigenvalue formulation (lines).

$n_{\text{iso}}^{\text{num}} = 5.64$ in the isotropic limit ($\langle S^2 \rangle = 1/2$) to $n_{\text{pc}}^{\text{num}} = 6.17$ in the perfect-cross limit ($\langle S^2 \rangle = 1$), corresponding to an increase of approximately 9%. These values differ slightly from the values 5.68 and 6.26 reported in Ref. [15], where the threshold was identified from the intersection of percolation-probability curves for different domain sizes Λ . Despite being generated from three distinct PDF families, the simulated thresholds approximately collapse onto a common curve when plotted against $\langle S^2 \rangle$. Within the class of distributions considered here, $\langle S^2 \rangle$ therefore provides an effective first descriptor of the percolation threshold.

Figure 4 compares the numerical simulations (Sec. IIC) with the eigenvalue formulation [Eq. (10)] predictions after rescaling each dataset according to Eq. (15). The abscissa $\langle S^2 \rangle / \langle S^2 \rangle_{\text{iso}}$ measures the departure from isotropy. At the isotropic and perfectly cross-aligned limits, the eigenvalue and fully preaveraged formulations, Eqs. (10) and (12), give identical thresholds: $n_{\text{iso}}^{\text{eig}} = n_{\text{iso}}^{\text{pre}} = \pi/2$ and $n_{\text{pc}}^{\text{eig}} = n_{\text{pc}}^{\text{pre}} = 2$. These theoretical values underestimate the simulated thresholds $n_{\text{iso}}^{\text{num}}$ and $n_{\text{pc}}^{\text{num}}$ by factors of approximately 3.6 in the isotropic limit and 3.1 in the perfectly cross-aligned limit. They also imply an increase of approximately 27% between the two limits. This systematic discrepancy arises from the omission of closed loops, which is the only structural approximation remaining in the eigenvalue formulation for the systems considered here. After rescaling (15), however, the simulation data closely follow the eigenvalue predictions for all three PDFs.

B. Dominant role of the averaged excluded area

Figure 5 quantifies the effect of retaining the orientation-resolved contact numbers by comparing the threshold from the eigenvalue formulation, Eq. (10), with that from the fully preaveraged approximation, Eq. (12). For all three PDFs, the normalized difference shown in Fig. 5 remains below approximately 1.2×10^{-3} , corresponding to less than 0.25% relative to the mean of the two thresholds. Since the eigenvalue formulation retains the full orientation-resolved contact ker-

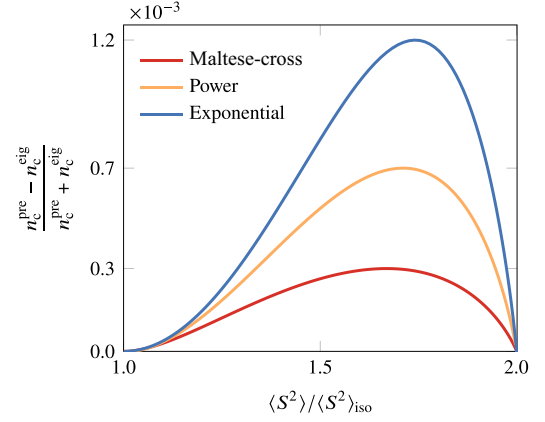


FIG. 5. The symmetric normalized difference $(n_c^{\text{pre}} - n_c^{\text{eig}}) / (n_c^{\text{pre}} + n_c^{\text{eig}})$ is shown as a function of $\langle S^2 \rangle / \langle S^2 \rangle_{\text{iso}}$ for the Maltese-cross, power, and exponential PDFs.

nel, whereas the preaveraged approximation replaces it by a single orientational average, the small difference between the two predictions indicates that orientation-dependent variations in the expected contact numbers provide only a secondary correction for the PDFs considered here. For the three PDF families considered, the fully preaveraged approximation predicts a threshold equal to or slightly higher than that obtained from the eigenvalue formulation. The magnitude and parameter dependence of the difference vary among the three PDFs. Thus, for equal-length penetrable sticks in two dimensions with $\langle S \rangle = 0$ and the symmetries in Eq. (3), the percolation threshold is governed primarily by the orientationally averaged excluded-area factor $\langle |\sin(\gamma_{ij})| \rangle$, or equivalently by the averaged excluded area $\langle a \rangle$ defined in Eq. (13).

Taken together with the agreement shown in Fig. 4 between the Monte Carlo results and the eigenvalue formulation, this finding indicates that the averaged excluded area captures the leading orientational dependence of the simulated thresholds.

C. Harmonic/moment interpretation of the averaged excluded area

The dominant role of $\langle |\sin(\gamma_{ij})| \rangle$ suggests a representation that makes the contribution of successive orientational moments explicit. Because the eigenvalue formulation closely follows the fully preaveraged approximation, this analysis can be based on the averaged excluded-area factor.

To express the fully preaveraged prediction in the normalized form used below, we substitute Eq. (12) into the definition of Y^{pre} in Eq. (16), obtaining

$$Y^{\text{pre}} = 2 \frac{2 - \pi \langle |\sin(\gamma_{ij})| \rangle}{4 - \pi}. \quad (17)$$

Evaluating the Fourier series of $|\sin(\gamma)|$ at the relative angle γ_{ij} gives

$$|\sin(\gamma_{ij})| = \frac{2}{\pi} \left(1 - 2 \sum_{k=1}^{\infty} \frac{\cos(2k \gamma_{ij})}{4k^2 - 1} \right).$$

For two independently sampled orientations, the relative angle is $\gamma_{ij} = \theta_j - \theta_i$. Averaging the Fourier representation over all

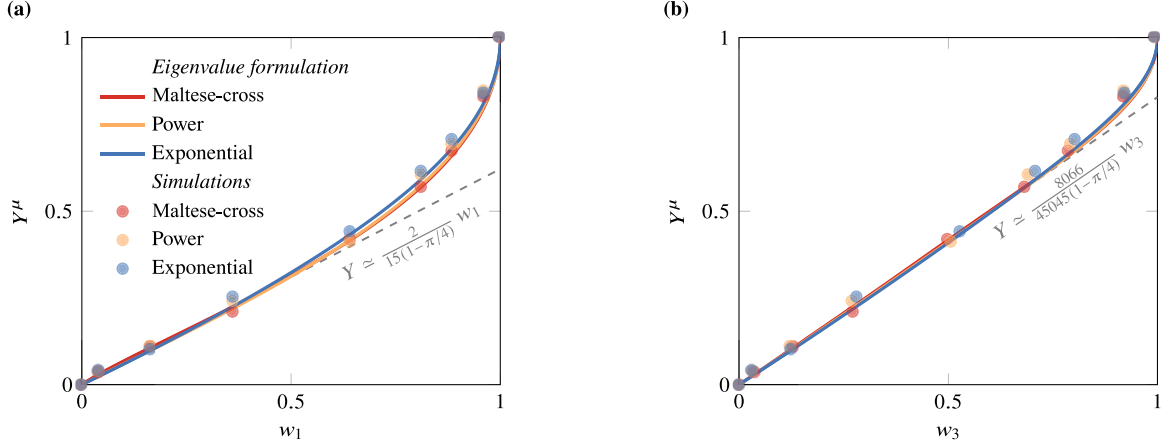


FIG. 6. The normalized threshold Y^μ defined in Eq. (16) is plotted against (a) w_1 defined in Eq. (21), obtained by retaining the first nonzero term in the harmonic expansion, and (b) w_3 defined in Eq. (22), obtained by retaining the first three nonzero terms. Symbols denote Y^{num} from the Monte Carlo simulations, solid curves denote Y^{eig} from the eigenvalue formulation, and black dashed lines show the linear approximations obtained by truncating the harmonic expansion after the first nonzero term in panel (a) and after the first three nonzero terms in panel (b).

orientation pairs requires evaluating

$$\langle \cos(2k\gamma_{ij}) \rangle = \langle \cos[2k(\theta_j - \theta_i)] \rangle.$$

Expanding the cosine of the angle difference and using the fact that θ_i and θ_j are independent and identically distributed yields

$$\langle \cos(2k\gamma_{ij}) \rangle = C_k^2 + D_k^2,$$

where

$$C_k = \langle \cos(2k\theta) \rangle, \quad D_k = \langle \sin(2k\theta) \rangle.$$

The reflection symmetry $\psi(\theta) = \psi(-\theta)$ gives $D_k = 0$ for every k . Moreover, using the π -periodic extension of the orientational PDF, $\psi(\theta) = \psi(\pi/2 - \theta)$ implies $C_k = (-1)^k C_k$, so that $C_k = 0$ for odd k . Consequently, only the terms with $k = 2q$ remain. Defining

$$X_q = C_{2q} = \langle \cos(4q\theta) \rangle, \quad (18)$$

the orientational average becomes

$$\langle |\sin(\gamma_{ij})| \rangle = \frac{2}{\pi} \left(1 - 2 \sum_{q=1}^{\infty} \frac{X_q^2}{16q^2 - 1} \right). \quad (19)$$

The first three coefficients, X_1 , X_2 , and X_3 , can be expressed in terms of moments of the order parameter S as follows:

$$X_1 = 2\langle S^2 \rangle - 1, \quad (20a)$$

$$X_2 = 8\langle S^4 \rangle - 8\langle S^2 \rangle + 1, \quad (20b)$$

$$X_3 = 32\langle S^6 \rangle - 48\langle S^4 \rangle + 18\langle S^2 \rangle - 1. \quad (20c)$$

Substituting the harmonic expansion in Eq. (19) into Eq. (17) allows the threshold variation to be expressed in terms of successive orientational harmonics. Retaining either (i) the first nonzero term in the summation, or (ii) the first three nonzero terms, defines the variables

$$w_1 = X_1^2, \quad (21)$$

and

$$w_3 = \frac{45\,045}{8066} \left(\frac{2}{15} X_1^2 + \frac{2}{63} X_2^2 + \frac{2}{143} X_3^2 \right). \quad (22)$$

Since X_1 varies from zero in the isotropic limit to one in the perfectly cross-aligned limit, $w_1 = X_1^2$ is already normalized. The prefactor in Eq. (22) is chosen so that w_3 has the same limiting values. Figure 6 plots the normalized threshold Y^μ against w_1 and w_3 . The agreement among the three PDF families improves when the first three harmonic terms are retained, showing that higher-order moments become increasingly relevant as the orientational PDF approaches perfect cross-alignment, where the exact expansion involves all even moments of S . For the three PDF families considered, the second moment captures the leading variation near isotropy, Fig. 6(a), whereas higher-order moments have their strongest influence near perfect cross-alignment. This behavior explains the high sensitivity of strongly cross-aligned systems to orientational perturbations. For example, 10% relative changes in $\langle S^2 \rangle$, from 1.0 to 0.9 near perfect cross-alignment and from 0.5 to 0.55 near isotropy, produce relative changes in n_c of 13.8% and 0.18%, respectively, for the Maltese-cross PDF in the eigenvalue formulation. Thus, even a small departure from perfect cross-alignment can substantially reduce the geometric percolation threshold of an idealized stick network.

IV. DISCUSSION AND CONCLUSIONS

The simulations show that orientational distributions having the same mean order parameter can nevertheless produce substantially different percolation thresholds. Although $\langle S \rangle = 0$ for all the distributions considered, the threshold increases from the isotropic to the perfectly cross-aligned limit as their higher-order orientational structure changes. This increase is markedly nonuniform: perturbations close to isotropy produce only small variations in the threshold, whereas comparable departures from perfect cross-alignment have a much stronger effect. A vanishing value of $\langle S \rangle$ is therefore insufficient to

comprehensively characterize geometric percolation within this class of distributions.

The comparison between the orientation-resolved eigenvalue formulation and the fully preaveraged approximation identifies the origin of this dependence. The eigenvalue formulation retains the full excluded-area kernel and the corresponding orientation dependence of the expected contact numbers, whereas the preaveraged approximation replaces the kernel with a single orientational average. Despite this simplification, the thresholds predicted by the two formulations differ by less than 0.25% relative to their mean for the three PDF families examined. Orientation-dependent variations in the expected contact numbers therefore provide only a minor correction, while the dominant orientational dependence is retained by the averaged excluded area $\langle a \rangle$ defined in Eq. (13).

The harmonic representation of the averaged excluded area further identifies the orientational information required in the different regimes. For the three distribution families considered, the leading variation close to isotropy is governed by the second moment of the orientational order parameter. Progressively higher-order moments become relevant as the distribution approaches perfect cross-alignment, where the exact expansion involves all even moments of S . This hierarchy explains the greater sensitivity of strongly cross-aligned networks to orientational perturbations. It also shows that the descriptor required to characterize the threshold depends on the regime: $\langle S^2 \rangle$ captures the leading variation close to isotropy, whereas higher-order information is required close to perfect cross-alignment.

The theoretical descriptions reproduce the normalized orientational trends more accurately than the absolute threshold values. The eigenvalue formulation underestimates the simulated thresholds by factors ranging from approximately 3.6 in the isotropic limit to 3.1 in the perfectly cross-aligned limit. It also predicts an increase of approximately 27% between the two limits, compared with approximately 9% in the simulations. This systematic discrepancy arises from the omission of closed loops, which is the only structural approximation of the eigenvalue formulation. The effective medium approximation derived in the Appendix retains the same inverse dependence on the averaged excluded-area factor while changing the prefactor. The orientational dependence and the absolute scale of the threshold should therefore be distinguished: the former is largely controlled by the averaged contact geometry, whereas the latter is more sensitive to the representation of network topology.

For real nanowire assemblies, geometric connectivity is only one of the factors governing device performance. Electrical conductivity and optical transmittance also depend on junction resistance, nanowire transport properties, and the spatial structure generated during deposition. Deposition strategies that promote cross-alignment may simultaneously alter nanowire spacing, affect the occurrence of locally dense or sparse regions [15], and improve junction quality [14]. The higher geometric percolation threshold associated with cross-alignment is therefore not inconsistent with the improved electrical or optical performance reported for some aligned networks [10–13]. Changes in spatial uniformity and transport properties may compensate for the increase in the geometric threshold caused by alignment.

For equal-length penetrable sticks and the symmetric orientational distributions examined, the averaged excluded area captures the dominant orientational dependence of the threshold, while the hierarchy of orientational moments explains why the threshold becomes increasingly sensitive as perfect cross-alignment is approached.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

APPENDIX: AN EFFECTIVE MEDIUM APPROXIMATION FOR THE PERCOLATION THRESHOLD

We apply an effective medium approximation (EMA) to evaluate the percolation threshold of widthless sticks randomly dispersed on a planar surface. To this end, we consider the related electrical-conductivity problem for a network generated by N penetrable and perfectly conducting sticks. Each node $i = 1, 2, \dots, N$ represents a stick. A bond with junction conductance $g_{ij} = g_0$ connects two intersecting sticks i and j , whereas $g_{ij} = 0$ when they do not intersect.

The EMA of Ref. [27] replaces the original network with a second network with the same nodes and identical effective bond conductances $\bar{g}$. This network is a complete graph with $N(N-1)/2$ bonds and an effective conductance $\bar{G} = N\bar{g}/2$ between any two nodes [28]. The value of $\bar{G}$ is determined by requiring the average two-node resistance of the original network to equal $1/\bar{G}$, which gives

$$\frac{1}{N} \left\langle \sum_{i \neq j} \frac{g_{ij}}{g_{ij} + \bar{G}} \right\rangle = 2, \quad (\text{A1})$$

where $\langle \cdot \rangle$ denotes the ensemble average over network configurations. The sum over $i \neq j$ is taken over ordered pairs and therefore counts each undirected bond twice.

For a generic pair of sticks i and j , we write

$$g_{ij} = g(\mathbf{R}_{ij}; \theta_i, \theta_j), \quad \mathbf{R}_{ij} = \mathbf{r}_i - \mathbf{r}_j,$$

where $\mathbf{R}_{ij}$ is the separation vector between their centers. Because positions and orientations are sampled independently, and all sticks follow the same spatial and orientational distributions, the ensemble average in Eq. (A1) can be written, in

the thermodynamic limit, as

$$\rho \int_{\mathbb{R}^2} \int_{-\pi/2}^{\pi/2} \int_{-\pi/2}^{\pi/2} \psi(\theta_i) \psi(\theta_j) \frac{g_{ij}}{g_{ij} + \bar{G}} d\theta_j d\theta_i d^2\mathbf{R}_{ij} = 2. \quad (\text{A2})$$

For nonparallel sticks, for which $\sin(\gamma_{ij}) \neq 0$, the vectors $\hat{\mathbf{u}}_i$ and $\hat{\mathbf{u}}_j$ form an oblique basis, and the relative-position vector can be written as

$$\mathbf{R}_{ij} = \xi_i \hat{\mathbf{u}}_i + \xi_j \hat{\mathbf{u}}_j,$$

where ξ_i and ξ_j are the corresponding coordinates [29]. The junction conductance is then

$$g(\mathbf{R}_{ij}; \theta_i, \theta_j) = \begin{cases} g_0, & |\xi_i| \leq L/2, |\xi_j| \leq L/2, \\ 0, & \text{otherwise.} \end{cases} \quad (\text{A3})$$

For nonparallel configurations, the coordinate transformation has Jacobian $d^2\mathbf{R}_{ij} = |\sin(\gamma_{ij})| d\xi_i d\xi_j$, where $\gamma_{ij} = \theta_j - \theta_i$. The parallel case follows by continuity: the excluded area between parallel widthless sticks is zero, so such configurations make no contribution to the spatial integral. Substitution of

Eq. (A3) into Eq. (A2), followed by integration over ξ_i and ξ_j , gives

$$n \langle |\sin(\gamma_{ij})| \rangle \left(\frac{g_0}{g_0 + \bar{G}} \right) = 2, \quad (\text{A4})$$

where $\langle |\sin(\gamma_{ij})| \rangle$ is the orientationally averaged excluded-area factor defined in Eq. (11).

The EMA percolation threshold is identified as the density at which $\bar{G}$ vanishes. Setting $\bar{G} = 0$ in Eq. (A4) yields

$$n_c = \frac{2}{\langle |\sin(\gamma_{ij})| \rangle}.$$

This result differs by a factor of two from Eq. (12), obtained from excluded-volume arguments and the Bethe-lattice approach of Ref. [18]. The two approximations differ in their treatment of network topology: closed loops are neglected in the Bethe-lattice approximation and represented approximately in the EMA. They consequently yield different prefactors while preserving the same dependence on the averaged excluded-area factor.

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