Anisotropic behavior in percolation of close-packed Janus disks

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Introduction.— The novelty of Janus particles [1], which have two surface areas of different properties, were appreciated very early in the field of soft matter [2]. Nowadays the particles can be synthesized by various methods and they could be used as surfactants, micro-motors, displays, catalysts, biosensors etc. [3–6]. The possibilities offered by Janus particles mainly depend on the fact that heterogeneous surfaces lead to anisotropic interactions. Harnessing the unique interactions, unconventional self-assembled structures of Janus particles can be made, such as gases of micelles [7, 8], entropy stabilized open crystals [9, 10], crystals of anisotropic orientational order [7, 11–25].

In most systems of Janus particles, positional and orientational motions are coupled, which leads to complex phase behavior. However, for close-packed crystals of Janus particles, positional vibrations are much less important as their contributions are very small. Thus close-packed Janus systems provide a platform where one can tune controlling parameters, e.g., the temperature or pressure, to explore orientational order mainly driven by orientation fluctuations [18–24, 26, 27]. Recently it was found that, in the orientationally ordered nematic phase of slender nanoparticles, the coupling between the particle density and the orientational order gives rise to interesting nonmonotonic behavior of percolation thresholds [28, 29]. At the percolation threshold, a system-spanning connected cluster first appears. Percolation deals with long-range connectivity and is one of the most applied models in statistical physics [30–32]. For anisotropic interacting Janus particles of isotropic shapes, eliminating positional vibrations associated with the particle density by close packing, we ask how orientation fluctuations affect connectivity behavior in crystal phases of orientational orders.

To answer the above question, in this Letter we explore connectivity of close-packed Janus disks in two dimensions (2D). We use a simple model of Janus disks on the triangular lattice, where particles interact with the Kern-Frenkel potential [33]. The model exhibits a continuous phase transition from a high-temperature disordered phase to a low-temperature orientationally ordered stripe phase [23]. Combining percolation theory and Monte Carlo (MC) simulation, we find that, in the stripe phase, though critical exponents are consistent with standard percolation, universal values of dimensionless quantities (e.g., Binder-like ratios and wrapping probabilities [34]) change continuously along the percolation line. Using theoretical results for wrapping probabilities of standard percolation in 2D [35–38], we get an effective aspect ratio $\rho_e$ to perform a spatial rescaling and relate quantitatively percolation in the orientationally ordered stripe phase to standard percolation. Thus the mechanism underlying the continuous variations of dimensionless quantities is that anisotropic interactions in the stripe phase change connectivity correlations to be anisotropic, but the anisotropic behavior can be captured by standard percolation. We also show that the mechanism can explain anisotropic percolation behavior in systems of aligned rigid rods [39–41]. Using anisotropic bond percolation [42] on the triangular lattice, we demonstrate by the isoradial-graph method [43–45] that a general relation between anisotropic percolation and standard percolation requires an effective shear transformation involving both the aspect ratio $\rho_e$ and an effective boundary twist $t_e$.

Model.— We consider a system of close-packed Janus disks with monodisperse patch sizes in 2D. Rhombus-shaped $L \times L$ triangular lattices with periodic boundary conditions are used and each vertex of the lattice is oc-
cupied by a disk with diameter one. To allow only orientational motion, the center of each Janus disk is fixed at a vertex of the lattice. The half-patch angle \( \theta \) (i.e., the Janus balance [46]) characterizes the size of the patch, as shown in Fig. 1. The Janus disks interact with a Kern-Frenkel potential [33]: two nearest-neighbor disks contribute an energy \(-\epsilon\) if the two patches on them touch each other, and, otherwise, they contribute a zero energy. When studying connectivity percolation, two disks are regarded as connected when they interact with an energy \(-\epsilon\). The unit of temperature is \( k_B T \), where \( k_B \) is the Boltzmann constant.

**FIG. 1.** Phase diagram of close-packed Janus disks in 2D. According to whether the system is ordered or percolated, the diagram is divided into four regions: an ordered and (not) percolated region, a disordered and (not) percolated region. The inset contains an enlargement near \( \theta/\pi = 0.503 \), as well as a snapshot of an ordered and percolated configuration. Vertical dashed lines indicate the position of \( \theta/\pi = 1/2 \) and the infinite-temperature percolation threshold \( \theta_p/\pi \approx 0.628 \) [47]. If not explicitly stated, lines are added to guide the eye, and the error bars are smaller than or comparable with the symbols for data points.

*Thermodynamic phase behavior.*— Figure 1 shows the phase diagram in the \( T - \theta \) plane. Previously it was found that, for \( 1/3 < \theta/\pi \leq 1/2 \), there is a continuous thermodynamic phase transition from a high-temperature disordered phase to a low-temperature orientationally ordered stripe phase [23]. For close-packed Janus particles in two-dimensional continuum space, preliminary results showed that this thermodynamic phase transition is still continuous [17]. Since at \( T = \infty \) the percolation threshold is \( \theta_p/\pi = 0.62776541(3) > 1/2 \) [47], to explore connectivity at finite \( T \), we need first understand thermodynamic phase behavior for \( \theta/\pi > 1/2 \). We performed extensive MC simulation of the above model using the Metropolis algorithm, and sampled same quantities as in Ref. [23]. We find that the continuous phase transition from the disordered phase to the stripe phase also exists for \( \theta/\pi > 1/2 \), and determine the phase transition line as shown in Figure 1 [38].

*Anisotropic percolation behavior.*— To explore connectivity of the Janus disks, we combine the critical polynomial method [48–53] with MC simulation. In the probabilistic geometric interpretation [51], for standard percolation in 2D, the critical polynomial is defined as \( P_B(\theta, L) \), for which \( R \) is the probability that there is a cross-wrapping cluster and \( R_0 \) is the probability of no wrapping. The root of \( P_B = 0 \) gives the percolation threshold when \( L \to \infty \). The critical polynomial has been demonstrated to be very powerful in determining percolation thresholds in 2D [47, 53, 54]. Wrapping probabilities were sampled in our MC simulation. Near the whole percolation line, it is found that, curves of \( P_B(\theta, L) \) cross and crossing points converge quickly to \( P_B = 0 \) and \( \theta_p \) as \( L \) increases. The fact \( P_B(\theta_p, L \to \infty) = 0 \) suggests that the percolation transition belongs to the universality class of standard percolation. We perform finite-size scaling analyses and find that critical exponents indeed take values for standard percolation [38]. The estimated percolation thresholds at different \( T \) are shown by blue triangles in Fig. 1. It is seen that, as \( T \) drops, the value of \( \theta_p \) decreases and it approaches \( \theta/\pi = 1/2 \) in the low-temperature limit. The orientationally ordered stripe phase is not guaranteed to be percolated.

**FIG. 2.** Critical wrapping probabilities along the percolation line of the Janus system: (a) Probability of wrapping in only one direction \( R_1 \) vs \( T \); (b) Wrapping probability \( R_2 \) vs \( T \). (c) Theoretical curve of \( R_2 \) vs the aspect ratio \( \rho \) for standard percolation on parallelogram-shaped triangular lattices. (d) The effective aspect ratio \( \rho_e \) vs \( T \) for the Janus system.

From theory of critical phenomena, scale invariance at the percolation threshold is related to the fact that many dimensionless quantities are independent of the system size, if finite-size corrections are neglected. Critical values of dimensionless quantities are “universal” in the sense that a quantity holds the same value for different lattices, short-range interactions, etc. For wrapping probabilities, along the percolation line in Fig. 1, we find that, in the disordered phase, they indeed take same values as those for standard percolation [36, 55]. However, in the orientationally ordered stripe phase \( (T < 0.24) \), we find that critical wrapping probabilities change continu-
ously as shown in Figs. 2(a) and 2(b). This implies that the orientational order affects “universal” critical properties, though it looks natural that, when \( T \) drops, the clusters are becoming easier to wrap around the direction parallel to the stripes (contributing most to \( R_1 \)) and more difficult to wrap around two directions (\( R_2 \)).

It has been known that many “universal” values of critical dimensionless quantities still depend on factors such as the system shape and boundary conditions [36, 37, 55–57], anisotropy of lattice couplings [58–64], statistical ensembles [34]. Thus we are interested in whether or how our results above could be typed into existing results for standard percolation. From the configurations near the percolation threshold in the stripe phase, it is seen that clusters are longer in the parallel direction than in the perpendicular direction, as exemplified in Fig. 3(a). After rescaling the parallel direction by a proper factor, the configuration would be isotropic, as shown in Fig. 3(b). The rescaling transforms an anisotropic system with size \( L \times L \) to an effective isotropic system with size \( L_{\parallel} \times L \) (\( L_{\parallel} < L \)). This leads us to hypothesise that anisotropic percolation of the \( L \times L \) Janus system is related to standard percolation with system size \( L_{\parallel} \times L \), i.e., with the aspect ratio \( \rho = (\sqrt{3}L)/ (2L_{\parallel}) \), and that the rescaling factor is \( q = L_{\parallel}/L = \sqrt{3}/(2\rho) \).

For standard percolation in 2D, values of critical wrapping probabilities depend only on \( \rho \) and the boundary twist \( t \) (being \( \rho/\sqrt{3} \) for the parallelogram-shaped triangular lattice), and their exact expressions are available [35–38], as illustrated for \( R_2 \) in Fig. 2(c). Thus one can compare numerical values of critical wrapping probabilities for the Janus system with theoretical values for standard percolation, to get an effective aspect ratio \( \rho_e \). In this way, we get the dependence of \( \rho_e \) on \( T \) for the Janus system, as shown in Fig. 2(d). While \( \rho_e \) is fixed at \( \sqrt{3}/2 \) in the disordered phase, it monotonically increases as \( T \) drops in the stripe phase (\( T < 0.24 \)), and should approach infinity in the low-temperature limit.

With \( \rho_e \) above, we test our hypothesis about the relation between percolation of the Janus system and standard percolation. First, from Fig. 3(a) to Fig. 3(b), \( q = \sqrt{3}/(2\rho_e) \) appears to be the proper rescaling factor. Second, we calculated the correlations \( g(r) \) (probabilities that two particles at a distance \( r \) belong to the same cluster) in both the parallel and perpendicular directions at \( \theta_p \). As shown in Fig. 3(c) for \( T = 0.23 \), the correlations of the Janus system can be collapsed into those of standard site percolation under proper rescaling [38]. Finally, we compare critical values of two dimensionless ratios at \( \rho = \rho_e \). The two ratios are related to cluster size distributions, and defined as: \( Q_1 = \langle C_i^2 \rangle / \langle C_i \rangle^2 \), \( Q_s = (3S^2 - 2S_i) / \langle S_i \rangle^2 \), where \( C_i \) (\( C_i \) for \( i \neq 1 \)) is the size of the largest cluster (other clusters) and \( S_l = \sum_i C_i^l \) is the \( l \)-th moment of cluster sizes. It can be seen from Figs. 3(d) and 3(e) that critical values of the two ratios for the Janus system are consistent with those of standard site percolation. The nonmonotonic behavior of \( Q_1 \) and the smooth approach of both ratios in the limit \( \rho \to \infty \) reflect evolution features of the probability density functions [57, 65]. Thus the Janus system is quantitatively related to standard percolation through the effective aspect ratio \( \rho_e \). Once \( \rho_e \) is determined, one can use results of standard percolation to predict universal behavior of the Janus system.

**FIG. 3.** Relations between percolation in the stripe phase of the Janus system and standard site percolation on the parallelogram-shaped triangular lattice. (a) A snapshot of the periodic Janus system, at \( \theta_p/\pi = 0.503027(2) \), with \( T = 0.23 \) and \( L = 256 \). The configuration is ordered in the parallel (horizontal) direction, as exemplified in the inset of Fig. 1. The dark blue region represents the largest wrapping cluster, with its holes (light yellow regions) being much longer in the parallel direction than in the perpendicular direction. (b) After rescaling the parallel direction by \( q = \sqrt{3}/(2\rho_e) \), with \( \rho_e \geq 2.85 \), the configuration becomes isotropic. (c) At parameters same as in (a), by proper rescaling, critical correlations of the Janus system in both the parallel (“para”) and perpendicular (“perp”) directions match those of site percolation on the triangular lattice with \( \rho \geq 2.85 \) (a parallelogram of size \( L_{\parallel} \times L = 88 \times 290 \)). The slopes of the curves agree with the theoretical value \(-5/24\) for standard percolation. (d) Critical dimensionless ratios \( Q_1 \) and \( Q_s \) of the Janus system are consistent with simulation results of site percolation when \( \rho = \rho_e \).

**FIG. 4.** Results for percolation of aligned rigid rods on periodic square lattices. (a) \( R_2 \) and \( \rho_e \) vs \( k \). (b) For different \( \rho = \rho_e \), values of \( Q_1 \) and \( Q_s \) are consistent with simulation results for site percolation on periodic square lattices.

**Application in other systems.** Beyond the Janus system, using \( \rho_e \), standard percolation can also be applied...
to explain anisotropic percolation behavior in other systems. For example, percolation of aligned rigid rods (also called $k$-mers as a rod consecutively occupies $k$ sites) were studied on square [39, 40] and triangular lattices [41], and it was found that critical percolation probabilities vary with changes in the rod length. Though it was realized that the variation comes from the system anisotropy, critical values different from those of isotropic systems were regarded as “nonuniversal” [39–41]. We performed MC simulation for aligned rigid rods of various sizes $k$ on periodic $L \times L$ square lattices. In the simulation [38], the system is treated as a random sequential adsorption, and wrapping probabilities are estimated at percolation thresholds determined by the critical polynomial method. By comparing critical values of $R_2$, as shown in Fig. 4(a), with the theoretical curve of $R_2$ for standard percolation on rectangular-shaped periodic square lattices ($t=0$) [35–38], we extract the dependence of $\rho_e$ on $k$, as plotted in Fig. 4(a), which suggests $\rho_e \approx 0.4k$ for large $k$. We also simulated standard site percolation on periodic square lattices of size $(pL) \times L$. When $p=p_0$, from Fig. 4(b), it can be seen that critical values of $Q_1$ and $Q_4$ for the system of aligned rigid rods nicely match those for site percolation.

$$\rho_e \approx \sin(\omega_2/2)\sin(\omega_1/2)$$

FIG. 5. Schematic plots of the isoradial graph for anisotropic bond percolation on the triangular lattice.

The anisotropic correlations in the stripe phase of the Janus system come from emergent orientational order, while those in the system of aligned rigid rods are introduced explicitly. For both models a spatial rescaling by $\rho_e$ is adequate to relate them to standard percolation. However, we find that for general anisotropic systems, an effective shear transformation described by $\rho_e$ and an effective boundary twist $\tau_e$ are needed. This is demonstrated in the following by studying anisotropic bond percolation on the triangular lattice. As shown in Fig. 5, in the original model, three edges of a triangle are occupied with different probabilities $p_0$, $p_1$ and $p_2$. At criticality the probabilities satisfy $p_0 + p_1 + p_2 - p_0 p_1 p_2 = 1$ [66]. Recently the method of isoradial graphs [43–45] was developed to prove the equivalence of critical exponents between anisotropic to isotropic systems, but the method has not been applied to dimensionless quantities such as wrapping probabilities. In isoradial graphs, for every edge, its length is adjusted to compensate its weight in order to make the model conformally invariant in the scaling limit [45]. Thus we expect that a critical dimensionless quantity takes the same value on different isoradial graphs of given $\rho$ and $t$. After the isoradial mapping, three vertices of a triangle locate on a circle, and the angles are determined by the Kenyon-Grimmert-Manolescu formula [43, 44] as

$$\omega_i = 3\arctan[\sqrt{3}(1-p_i)/(1+p_i)], \ i=0, 1, 2,$$

as illustrated in Fig. 5. Thus, given values of $p_0$ and $p_1$, the value of $p_2$ can be solved from the equation of critical condition, and the rhombus at the left side of Fig. 5 transforms to the parallelogram with $\alpha = \omega_2/2$ at the right side. For the original model on a $L \times L$ rhombus-shaped periodic triangular lattice, we derive that the graph after the isoradial mapping has $\rho_e = \sin(\omega_2/2)\sin(\omega_1/2)/\sin(\omega_0/2)$ and $\tau_e = \cos(\omega_2/2)\sin(\omega_1/2)/\sin(\omega_0/2)$. The isoradial mapping is an effective shear transformation. We have numerically verified that wrapping probabilities of anisotropic bond percolation on the triangular lattice are equal to theoretical values of standard percolation on periodic systems with aspect ratio $\rho_e$ and twist $\tau_e$ [38].

Summary and prospects.— In this Letter, we investigate connectivity percolation of close-packed Janus disks in 2D. The orientational order in the stripe phase leads to anisotropic percolation behavior, since correlation is stronger in the direction parallel to the stripes than in the perpendicular direction. The anisotropic behavior is reflected in the continuous variation of dimensionless quantities such as wrapping probabilities. We derive an effective aspect ratio $\rho_e$ and find that universal properties of the Janus system are consistent with those of standard percolation. We also apply $\rho_e$ to relate standard percolation to percolation of aligned rigid rods. In general an effective shear transformation with aspect ratio $\rho_e$ and an effective boundary twist $\tau_e$ are needed to relate anisotropic percolation to standard percolation, which we demonstrate by applying the isoradial-graph method to anisotropic bond percolation on the triangular lattice. The results are of importance in designing materials with tunable anisotropic connectivity-related properties such as conductivity [28, 67].

It was shown recently that an effective shear transformation also relates the anisotropic Ising model on a square to the isotropic Ising model on a parallelogram [63, 64], though different techniques were used to calculate effective parameters [68]. Bond percolation and the Ising model correspond to the $q=1$ and 2 Potts models [69], respectively. For the Potts model of other $q$ in the Kasteleyn-Fortuin representation in 2D, where exact wrapping probabilities are available [70, 71] and isoradial graphs have been studied [45], further research should uncover similar physics.

Finally, we anticipate that the mechanism is valid for other boundary conditions, in higher dimensions and continuum space [28], as long as correlations are weakly anisotropic [63]. Strongly anisotropic percolation systems [72] also deserve more attention.

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[38] See Supplemental Material at [URL will be inserted by publisher] for: (1) Details for the system of close-packed Janus disks, including those for the thermodynamic phase transition and those for the percolation transition. (2) Details for the system of aligned rigid rods. (3) Details for anisotropic bond percolation, which verify the theoretical results obtained using the method of isoradial graphs. (4) A script for calculating exact values of wrapping probabilities in 2D using expressions from the literature.
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