Explosive percolation in a nanotube-based system

Yup Kim, Yeo-kwang Yun, and Soon-Hyung Yook*
Department of Physics and Research Institute for Basic Sciences, Kyung Hee University, Seoul 130-701, Korea
(Received 2 July 2010; revised manuscript received 25 August 2010; published 3 December 2010)

Using the percolation theory, we study the underlying mechanism in the formation of single-walled nanotube bundles with uniform diameter. By applying the cluster repulsion process to stick percolation, we find that the transition becomes explosive. To understand the transition nature, we first investigate the scaling behavior of transition interval Δ. By comparing the results with loopless and loop-allowed bond percolations, we find that the loops crucially affect the scaling behavior of Δ, and Δ is not universal. Moreover, the scaling behavior of Δ for the present nanostick systems is the same as that for loopless bond percolation. For more systematic studies on the transition nature, we also measure the changes in order parameter during the stick removal process and show that there exists a hysteresis. The results more clearly show that the transition of the stick system with cluster repulsion is discontinuous.

DOI: 10.1103/PhysRevE.82.061105 PACS number(s): 64.60.ah, 05.45.Df, 72.80.Tm, 73.63.Fg

I. INTRODUCTION

Recent developments of thin film of nanowires, such as silicon nanowires [1] and carbon nanotubes [2–4], provide enormous applications in electronics, optoelectronics, and sensors. Especially, the studies on the changes in electrical transport in composites of conducting fibers, such as functional organic materials and nanotubes, have become of prime importance in designing nanodevices [5–7]. In such systems, it is well known that the addition of small number of nanotubes substantially modifies their transport properties without the modification of mechanical properties [8–13]. Especially, the transition from an insulator to a conductor in the composites has been one of the well-known phenomena in such systems and plays an important role in the nanodevice designing. This transition is generally characterized by using a percolation theory [14]. The emergence of percolating paths in nanotube composites and understanding their electrical properties are crucial in designing a better nanodevice [5–7].

In many experimental studies, single-walled nanotubes (SWNTs) with large aspect ratio have been shown to play an important role in designing nanoscale devices [2–4]. Such nanotubes with large aspect ratios can be well approximated by widthless sticks [5,6]. Therefore, the understanding of the percolation of a stick system is very important to understand the underlying mechanism in nanoscale systems. The early studies on the percolation of fiber or nanotube composites have been focused on the finding of critical length of nanotubes, ℓ_c, by which an infinitely connected cluster emerges when the total number of sticks is fixed [15,16]. The geometrical properties of stick percolation are known to belong to the same universality class with the ordinary percolation (OP) [17,18], while the dynamical exponent can be different from that of ordinary percolation [19].

Since the suggestion of Achlioptas *et al.* of a model which shows an explosive transition, there have been many attempts to understand the general properties and conditions which cause such transition [20–24]. Achlioptas *et al.* originally introduced the model for the growth of random networks and showed that the order parameter discontinuously jumps to 1 when the growth probability of large clusters is suppressed. More recently, Ziff showed that the loopless bond percolation with Achlioptas' process (AP) in two dimensions ($2d$) also undergoes such discontinuous transition [21]. These indicate that the inhibition of the growth of large clusters produces clusters having almost the same size. An example of such transition was found in the human protein homology network [25].

A more interesting example of such discontinuous transition is found in the nanotube systems. Laser-vaporized method [26] and electric-arc technique [27] produce close-packed SWNTs with uniform diameters. Based on the percolation theory [14], the bundle size should not be uniform without the suppression of large bundle growth. Therefore, the net effective growth mechanism for bundle formation of SWNTs with uniform diameters can be well approximated by the suppression of the coagulation between large bundles, like in the explosive percolation transition. Thus, understanding the growth mechanism which causes clusters of uniform size is very important to produce SWNT bundles from the practical point of view. In this paper, therefore, we study the effect of cluster repulsion of nanotubes and show that the cluster repulsion in the considered theoretical model causes a first-order transition by measuring the hysteresis. Moreover, in the ordinary bond percolation, it is natural to allow the formation of loops, while the original explosive percolation model does not allow any loop. As we will show later, the scaling behavior of transition interval is crucially affected by loops [20]. By showing that the transition interval in stick systems can reproduce the same scaling behavior with loopless $2d$ bond percolations [21], we find that the size of loops is quite small compared to the entire system size at the transition threshold in the nanostick systems. Thus, we find that the nanostick systems can be effectively regarded as the loopless system with the inhibition of the growth of large clusters.

*syook@khu.ac.kr

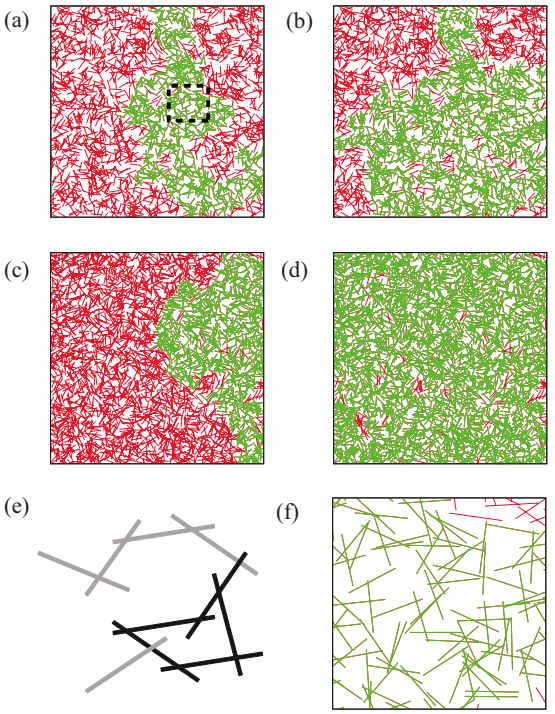

FIG. 1. (Color online) (a) and (b) show the snapshots of the largest cluster for OP. (c) and (d) show those for AP. (a) and (c) are the snapshots at the threshold of percolation transition. (b) and (d) are the snapshots when 200 sticks are added to (a) and (c), respectively. The green (light gray) sticks denote those that form the spanning (largest) cluster. Red (dark gray) sticks are those in the small clusters. (e) Schematic diagram for a loop. Black sticks are connected back to themselves which form a closed loop. (f) Enlarged plot of the dashed square in (a).

II. EXPLOSIVE PERCOLATION

A. Model

The AP in the nanostick system [5,6] is defined as follows: (I) We place two sticks α and β at random positions with random orientations. (II) Let $\{s_{\alpha_1}, s_{\alpha_2}, \ldots, s_{\alpha_n}\}$ ($\{s_{\beta_1}, s_{\beta_2}, \ldots, s_{\beta_m}\}$) be the sizes of clusters which form into a new big cluster with the size $\Sigma_{k=1}^{n} s_{\alpha_k}+1$ ($\Sigma_{k=1}^{m} s_{\beta_k}+1$) by placing stick α (β). Here, the cluster size is defined by the number of sticks in the cluster. Then calculate the products

$$\pi_\alpha = \prod_{i=1}^{n} s_{\alpha_i}, \quad \pi_\beta = \prod_{j=1}^{m} s_{\beta_j}. \tag{1}$$

This rule is generally called product rule. (III) If $\pi_\alpha \leq \pi_\beta$ ($\pi_\alpha \geq \pi_\beta$) then stick β (α) is removed. Processes (II) and (III) prefer the connection between small clusters, which causes the cluster repulsion or suppresses the growth of large clusters. In the following simulations, we use a two-dimensional square of linear sizes $L=5-40$. The length of each stick, ℓ, is set to $\ell=1$ ($\ll L$).

For a qualitative description, we compare the snapshot of the largest clusters in the OP and that under AP in Fig. 1. Figures 1(a) and 1(c) show the snapshots when the spanning cluster emerges, for OP and AP, respectively. Figures 1(b) and 1(d) show the configurations of the largest cluster when 200 more sticks are added to the configurations in Figs. 1(a) and 1(c). The snapshots clearly show that, once the spanning cluster emerges, the size of the largest cluster under AP grows much faster than that of OP when the same number of sticks is added. Therefore, the transition under AP is more explosive than OP. In Fig. 1(e) we show a schematic for a definition of a loop. The black sticks in Fig. 1(e) are connected back to themselves, which form a closed loop. As we shall show, for $2d$ percolations, the existence of loop affects the scaling behavior of transition interval. In Fig. 1(f) we enlarge the dashed square in Fig. 1(a). This enlarged configuration shows that the size of loops in the largest cluster is negligibly small compared to L. Thus, the transition becomes quite similar to the loopless $2d$ explosive percolation transition [21]. The effect of the loops will be discussed later in detail.

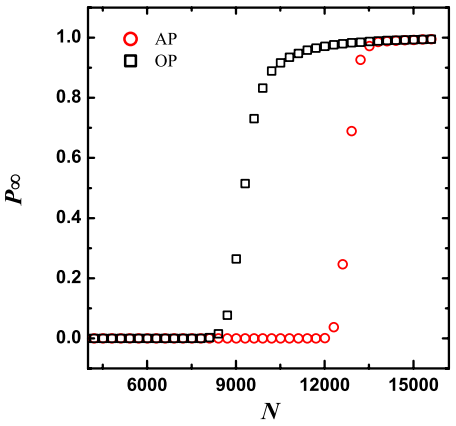

FIG. 2. (Color online) Plot of P_∞ against N when $L=40$. The (black) squares represent P_∞ for the OP and the (red) circles denote P_∞ under AP.

B. Order parameter

For a quantitative measurement of the changes in geometrical properties of stick system, we measure the order parameter. The order parameter P_∞ is defined by the probability that a stick belongs to a spanning (largest) cluster [14,18]:

$$P_\infty = \frac{N_\infty}{N}. \tag{2}$$

Here, N_∞ is the number of sticks in the spanning cluster and N is the total number of sticks placed in a given square. Figure 2 shows the comparison of P_∞ between OP and AP when $L=40$. The data in Fig. 2 show that the AP delays the transition as observed in the $2d$ loopless bond percolation [21] or in complex networks [20]. Moreover, as shown in the data, the transition under AP is more abrupt than that of the OP. This abrupt transition provides evidence that AP causes a first-order transition.

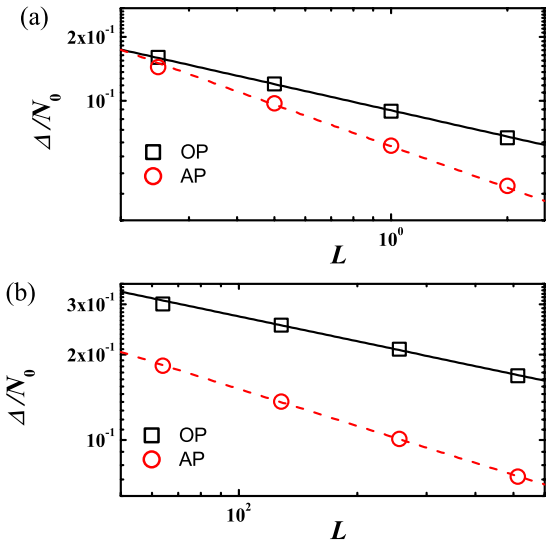

FIG. 3. (Color online) Δ/N_0 for various L's. (a) The (black) squares represent the data obtained from the ordinary stick percolation. The (red) circles display the data for the stick systems with AP. The (black) solid line represents the relation $\Delta/N_0 \sim L^{0.40}$ and the (red) dashed line denotes $\Delta/N_0 \sim L^{0.65}$. (b) Δ/N_0 for lattice bond percolation, in which the formation of loops is allowed. In the ordinary bond percolation we obtain $\Delta/N_0 \sim L^{0.29}$ [solid line in (b)]. For AP with loops in the lattice percolation, we obtain $\Delta/N_0 \sim L^{0.43}$ [dashed line in (b)].

C. Transition interval: Comparison of stick percolation with 2*d* bond percolation

To distinguish the transition property under AP from that of OP, a new method was suggested in [20]. The main idea of the suggested method consists of the investigation of the width of the transition interval. In order to apply the method to our stick system, we define the minimum number of sticks, N_0, at which P_∞ becomes 1. We also define N_1, where P_∞ equals $N_0^{-1/2}$, and N_2 at which $P_\infty = 1/2$. Then we define the width of the transition interval as $\Delta = N_2 - N_1$. In $2d$ loopless bond percolation, Δ is known to satisfy a power law $\Delta/N_0 \sim L^{-\delta}$ both for OP and AP. For OP, δ is closely related to the typical cluster mass s^* which scales as $s^* \sim |N - N_c|^{-1/\sigma}$ [21]. Here, N_c represents the critical stick number which corresponds to the percolation threshold. From a simple scaling argument, δ can be estimated as $\delta = \sigma/2 = 36/91$ for loopless $2d$ OP [21]. As shown in Fig. 3(a), δ for ordinary stick percolation also agrees very well with the results in loopless ordinary bond percolation. On the other hand, the numerical value of δ for AP is known as $\delta \approx 0.68$ for a $2d$ loopless case. The data in Fig. 3(a) also show that the measured value of δ for AP agrees very well with that of the loopless bond percolation under AP [21].

Since the geometrical properties of continuum percolation are identical to those of bond (or site) percolation [14,18], we consider ordinary bond percolation to study the effect of loops on the scaling behavior of Δ. As shown in Fig. 3(b), we find that the value of δ dramatically changes when loops are allowed. From the data in Fig. 3(b) we obtain $\delta \approx 0.29$ for ordinary bond percolation. This value is significantly smaller than that obtained from the ordinary stick percolation or from the loopless OP [21]. This indicates that the scaling arguments near the critical point suggested by Ziff [21] seem to be valid only for the loopless percolation and for our stick system. Moreover, this implies that our stick system has no effective loops in the global scale as observed in Fig. 1(f). When we apply AP to the lattice system with loops, we find $\delta \approx 0.43$ [see the dashed line in Fig. 3(b)].

For OP [see the squares in Figs. 3(a) and 3(b)], the difference in δ between the loopless bond percolation and loop-allowed bond percolation can be understood from a simple scaling argument. In contrast to the loopless bond percolation, the existence of loops allows us to connect the sites inside the same cluster. Thus, more occupied bonds are required to produce a macroscopically large cluster, and this increases the percolation threshold; Δ should be rescaled in a nontrivial way. It is well known that the clusters at the percolation threshold form a ramified fractal [14]. For the loopless case, the newly added bond always connects two different clusters. The probability to find such bond is proportional to the surface area of the ramified fractal, i.e., $\xi^{d_f} \sim L^{d_f}$ [14]. Here, ξ is the correlation length. On the other hand, for the loop-allowed bond percolation, any empty bond can be selected and the probability to find such bond is simply proportional to L^d [14]. Thus, the ratio between the intraconnection and interconnection probabilities becomes L^{d-d_f}. This implies that the occupation probability of loop-allowed bond percolation is over estimated by the factor L^{d-d_f} compared to that of the loopless bond percolation. Therefore, $\Delta_{loopless} = \Delta_{loop\text{-}allowed} L^{d_f - d}$. Since $d_f \simeq 1.9$ for $d=2$, $\delta_{loopless} \simeq \delta_{loop\text{-}allowed} - 0.1$, which agrees very well with the obtained δ in Figs. 3(a) and 3(b). This clearly shows that the loops crucially affect the scaling behavior of Δ. Thus, when one considers nanostick systems in which the effect of loops can be ignored, our stick systems can be used as a model system for further studies.

D. Hysteresis

Note that the measurements of Δ for stick percolation and loop-allowed bond percolation show that the δ is not universal. The results in Fig. 3 indicate that the behavior of Δ strongly depends on geometrical properties. Therefore, the limiting behavior of Δ/N_0 is not sufficient to distinguish the explosive percolation from OP. One of the most generally accepted and the simplest methods to verify whether the observed transition is first order or not is the measurement of the hysteresis [28,29]. The hysteresis is a history-dependent property of a system and is usually observed in the first-order phase transition because of the metastable state. If the transition is first order then the route of changes in P_∞ during stick addition process would be different from that for stick removal process. In Fig. 4, to check the existence of hysteresis we compare the measured P_∞'s when we add the sticks (black squares) and when we remove the sticks (red circles) for $L=40$ and $L=10$ (see the inset). For the removal process, we slightly modify rule (III) to easily break the larger clusters, since rules (II) and (III) suppress the formation of a large cluster, i.e., if $\pi_\alpha \geq \pi_\beta$ then we remove stick α. With this modified rule we find that there exists a hysteresis for

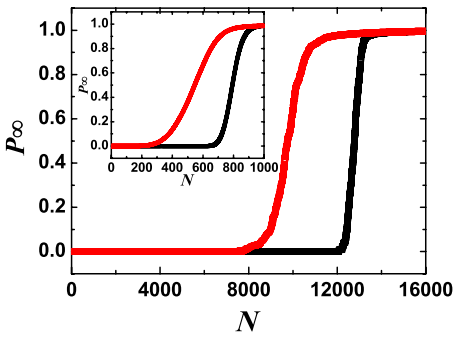

FIG. 4. (Color online) Hysteresis of P_∞ for $L=40$. Black squares represent P_∞ when we add sticks and red (gray) circles denote P_∞ when we remove sticks. Inset: the same plot for $L=10$.

any L as shown in Fig. 4. This clearly shows that the transition is first order.

III. SUMMARY

In summary, we study the explosive percolation of nanostick system. By imposing AP to the stick percolation we find that the transition is delayed and becomes more abrupt than that of OP. From the measurement of Δ we show that Δ of stick percolation with AP satisfies a power law, $\Delta/N_0 \sim L^{-\delta}$, as in $2d$ loopless bond percolation under AP [21]. However, by investigating Δ for OP, we find that δ depends on the existence of loops; thus, it is not universal. For more systematic analysis on the transition nature, we measure P_∞ during the stick removal process and observe the hysteresis. This clearly shows that the AP causes a first-order transition. Moreover, since the formation of nanotube bundles and stick percolation with AP share the same growth mechanism, i.e., suppressing the coagulation of different bundles or clusters, we expect that the model studied in this paper can provide a theoretical framework for the future studies on various physical properties of nanotube bundle formation.

ACKNOWLEDGMENTS

This work was supported by the National Research Foundation of Korea Grant funded by the Korean Government (2009-0073939), by the Mid-career Research Program through NRF grant (MEST) (Grant No. 2009-0052659), and by the Korea Research Foundation grant funded by the Korean Government (MOEHRD, Basic Research Promotion Fund) (KRF-2007-313-C00279).

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