

Conductivity of Stick Percolation Clusters with Anisotropic Alignments

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The conductivity of random resistor networks composed from percolating clusters of two-dimensional (2D) stick systems with anisotropic alignments is analyzed by using a finite-size scaling analysis for comparison to the conductivity of single-walled carbon-nanotube bundle film networks. For the conductivity analysis, we first calculate the critical properties of the percolation transition of 2D stick systems with anisotropic alignments. Even though the percolation transition stick density increases rapidly as the anisotropy is enhanced, the conductivity and the critical properties hardly vary. The resultant conductivity exponent of the stick networks at the percolation threshold is nearly the same as that of the lattice critical percolation clusters regardless of the anisotropy and the resistance ratio $r = R_{jct}/R_{NT}$, where R_{jct} is the stick-to-stick junction resistance and R_{NT} is the resistance of a stick.

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I. INTRODUCTION

Single-walled carbon-nanotubes (SWNTs) with large aspect ratios have drawn many studies because of their important role in designing nanoscale devices [1–3]. Recently the electrical conductivity of SWNT networks has been studied experimentally [4,5] and theoretically [5–8]. Especially Du *et al.* [5] studied the conductivity dependence of nanotube/polymer composite film networks on the alignment and the concentration of such nanotubes, and found a strong dependence of the percolation properties on the anisotropy of the alignments. They still argued that the conductivity critical exponent depends on the anisotropy. Hecht *et al.* [4] experimentally studied the dependence of the conductivity σ of the SWNT film networks on the SWNT bundle length ℓ as

$$\sigma \sim \ell^{1.46}. \quad (1)$$

This power-law dependence of the conductivity on ℓ was qualitatively explained based on the fact that the resistance along the SWNT bundle itself (R_{NT}) is much smaller than the resistance due to tube-to-tube junction (R_{jct}) or $R_{jct} \gg R_{NT}$ [4].

A good theoretical model for SWNT film networks is a two-dimensional (2D) stick system, which is one of the well-known continuum percolation models [4, 5, 7–11]. Classical works on the 2D stick percolation system [9] showed that the universality of the percolation transition of isotropic stick systems is the same as that of

the lattice percolation transition. For this, Balberg *et al.* [9] have calculated the critical exponents of the percolation transition in the infinite-size or thermodynamic limit. They put N sticks with the length ℓ in a unit square [9]. By using the fact that the critical percolation thresholds N_c or ℓ_c satisfy the relation $\ell^2 N_c = \ell_c^2 N$, they estimated the critical exponents from the relations

$$N_p/N \simeq (N/N_c - 1)^\beta = (\ell^2/\ell_c^2 - 1)^\beta, \quad (2)$$

$$S(p) \simeq (N/N_c - 1)^\gamma = (\ell^2/\ell_c^2 - 1)^\gamma, \quad (3)$$

$$\sigma \simeq (N/N_c - 1)^t = (\ell^2/\ell_c^2 - 1)^t, \quad (4)$$

where N_p is the number of the sticks in the percolating cluster, $S(p)$ is the average size of finite clusters and σ is the conductivity of the resistor networks composed from the percolating cluster. Because $(N/N_c - 1)$ or $(\ell^2/\ell_c^2 - 1)$ directly corresponds to $(p - p_c)/p$ for a lattice percolation system [12], Balberg *et al.* [9] obtained the critical exponents as $\beta = 0.14$, $\gamma = 2.3$, and $t = 1.24$ without the modern finite-size scaling analysis [12–14]. Balberg *et al.* [9] also studied the dependence of N_c on the alignment of the sticks. However, for comparison of the experimentally-found result (1) to σ for stick percolation networks, one needs a finite-size scaling analysis, because σ in a finite-size system should satisfy the relation $\sigma \sim \ell^{t/\nu}$ with the correlation length exponent ν . In this sense, recently, finite-size scaling analyses were performed for stick percolation systems [7,8]. In the finite-size scaling analysis [7], the percolation transition of the isotropic stick system was shown to belong to the same universality class as the lattice percolation transition. Furthermore, the finite-size scaling analysis [8] ar-

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gued that the conductivity exponent t or t/ν for such stick critical percolation clusters does not depend on the ratio $r = R_{jct}/R_{NT}$ and obtained $t = 1.28$ ($t/\nu = 0.96$) regardless of the value r . If the finite-size scaling analysis is right, the simple 2D stick percolation system cannot explain the experimental result (1) with $t/\nu \simeq 1.46$. The discrepancy might come from the anisotropic effects of the stick alignment. The dependence of the percolation threshold and the conductivity on the anisotropy of the stick alignments was studied by Du *et al.* [5] for comparison to the experimental result, but the dependence of t or t/ν on the anisotropy is not quantitatively and clearly understood.

In this paper we use a careful finite-size analysis of the 2D stick percolation system to study the effects of the anisotropy of the stick alignments and the ratio $r = R_{jct}/R_{NT}$ on the critical properties and the conductivity of 2D stick percolation systems. As we shall see, the critical properties and t/ν do not depend on the anisotropy, and the difference of t/ν in the 2D stick systems from the experimental value in Eq. (1) should originate from some other physical factors.

This paper is organized as follows. In Sec. II, the 2D stick model is explained, with careful definitions of various quantities and relations. Section III, a finite-size scaling analysis for the critical properties of the percolation transition is carried out considering the effects of anisotropic alignments. In Sec. IV, σ of the random resistor networks composed from the spanning clusters with a finite value of r and the effects of anisotropic alignments are analyzed. In the final section, the summary and relevant discussions are presented.

II. TWO-DIMENSIONAL STICK PERCOLATION SYSTEM

One can think of two kinds of equivalent models for the 2D stick percolation system. The first kind of model, which we call model **A**, uses N sticks of lengths ℓ (< 1) distributed randomly on a 2D unit square. In the other kind of model, model **B**, N sticks of the unit length ($\ell = 1$) are randomly distributed on a square with a side of length L . Model **A** can be exactly mapped with a one-to-one correspondence to the model **B** by multiplying both the stick length and the length of the side in model **A** by ℓ^{-1} ($= L$). Therefore, the physical properties of the percolation transition using model **A** are the same as those of model **B**. For convenience, we use model **B** from now on.

A typical configuration in model **B** is shown in Fig. 1. The position and the orientation of a stick i are identified by the coordinate of the center, (x_i, y_i) , and the orientation angle, θ_i , with respect to the y -axis. If sticks are placed randomly with $-\pi/2 \leq \theta_i \leq \pi/2$, then the configuration of sticks is isotropic. If the condition $-\theta_{cut} \leq \theta_i \leq \theta_{cut}$ with $\theta_{cut} < \pi/2$ is imposed, then

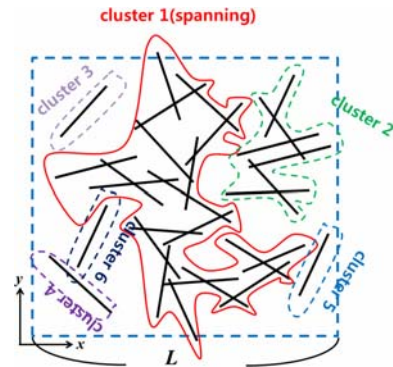


Fig. 1. (Color online) Typical configuration in model **B**. The length of a side of the square is L ($= \ell^{-1}$). Clusters of sticks are shown. Because a spanning cluster exists, the configuration is one with a density of sticks larger than the percolation threshold density m_c .

the configuration should have anisotropy. In the limit $\theta_{cut} \rightarrow 0$, the configuration approaches that in which all the sticks are parallel to one another. If a pair of sticks cross each other, the pair belongs to the same cluster as shown in Fig. 1. The size of a cluster is defined by the number of sticks that belong to the cluster. If the density of sticks or m ($= N/L^2$) increases, a cluster that connects the top side of $y = L$ to the bottom side of $y = 0$ appears. The connecting cluster is called a spanning cluster or a percolation cluster. The percolation transition threshold density m_c is the density at which at least one percolating cluster exists if $m > m_c$ ($= N_c/L^2$) in the limit $L \rightarrow \infty$.

III. FINITE-SIZE SCALING ANALYSIS OF THE PERCOLATION TRANSITION WITH ANISOTROPY

As introduced, the **finite-size scaling analysis (FSSA)** for the percolation transition with $\theta_{cut} = \pi$ or for the isotropic alignments was carried out to confirm that the transition of 2D isotropic stick systems belongs to the same universality class as the lattice percolation transition [7], as Balberg *et al.* [9] showed by using an infinite-size analysis. In this section, we want to check that the transition of anisotropic stick systems belongs to the same universality class as the lattice percolation transition by use of a FSSA. Even though several works have studied the dependence of the transition threshold density m_c on θ_{cut} [5,10], the dependence of the universality of the transition on θ_{cut} has barely been investigated. Because the dependence of the correlation length exponent ν on θ_{cut} is needed to estimate the dependence of the conductivity exponent t on θ_{cut} in the next section, calculating the dependence of the critical exponents on θ_{cut} by using the FSSA is important. For the FSSA of the stick model, the simulation data are obtained by

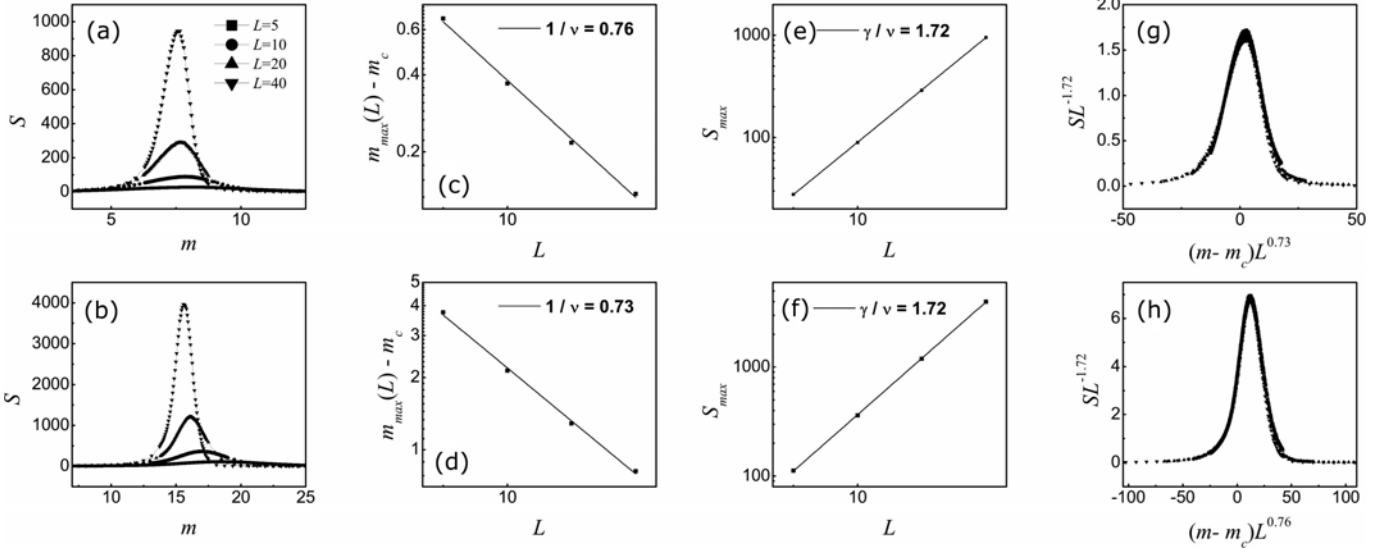


Fig. 2. Average size S of finite clusters against the density m for various L with (a) $\theta_{cut} = \pi/4$ and (b) $\theta_{cut} = \pi/9$. (c) The line with the value of $1/\nu$ is the fit of $m_{max}(L)$ to Eq. (6) for $\theta_{cut} = \pi/4$. (d) The same fit as (c) for $\theta_{cut} = \pi/9$. (e) The line with the value of γ/ν is the fit of S_{max} to Eq. (7) for $\theta_{cut} = \pi/4$. (f) The same fit as (e) for $\theta_{cut} = \pi/9$. (g) The data collapse to the scaling function in Eq. (5) by use of the exponents obtained in (c) and (e) for $\theta_{cut} = \pi/4$. (h) The same collapse as (g) by use of the exponents in (d) and (f) for $\theta_{cut} = \pi/9$.

averaging over $10^5 \sim 10^6$ randomly-generated configurations of model B for each set of m and L . Figures 2(a) and (b) show the data obtained for the average size S of finite clusters, which depends on the system size L and the stick density m . The finite-size scaling ansatz of S for $m \simeq m_c$ or the critical region [12] can be written as

$$S(m, L) = L^{\gamma/\nu} f((m - m_c)L^{1/\nu}), \quad (5)$$

where m_c is the critical density of sticks, which depends on θ_{cut} ; *i.e.*, $m_c = m_c(\theta_{cut})$. The scaling function f shows the scaling behavior of $f(x) \sim x^{-\gamma}$ for $x \gg 1$ and $f(x) = const.$ for $x \ll 1$. Furthermore the maximal value $S_{max}(L)$ of S and the density $m_{max}(L)$ at which S becomes S_{max} have the following properties:

$$m_{max}(L) = m_c + aL^{-1/\nu} \quad (6)$$

and

$$S_{max}(L) \sim L^{\gamma/\nu}. \quad (7)$$

From Eq. (7) and the data as in Figs. 2(a) and (b), we estimate $m_c(\theta_{cut})$ and the correlation length exponent $\nu(\theta_{cut})$. The fittings of the relation (7) to the data for $\theta_{cut} = \pi/4$ and $\theta_{cut} = \pi/9$ are shown in Figs. 2(c) and (d), respectively. From such fittings, $m_c(\theta_{cut})$ are estimated as in Fig. 3. We reconfirm the relation $m_c(\pi/2)\ell^2 = m_c(\pi/2) = 5.637(2)$ with $\ell = 1$ for the isotropic case [7, 15], as shown in Fig. 3. The critical density m_c does not vary much for the interval $5\pi/18 < \theta_{cut} \leq \pi/2$, but rapidly changes for the interval $\theta_{cut} < 5\pi/18$ as $m_c \simeq \theta_{cut}^{-0.9}$. This behavior of $m_c(\theta_{cut})$ based on the FSSA is qualitatively very similar to that from

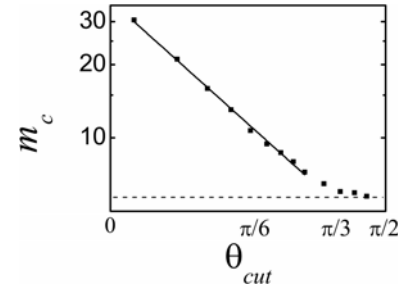


Fig. 3. Threshold density $m_c(\theta_{cut})$ against θ_{cut} . The solid line denotes the relation $m_c \sim \theta_{cut}^{-0.9}$. The dashed line corresponds to $m_c = 5.637$

an experimental analysis [5] and that from the infinite-size analysis for the critical density [10]. In contrast to the rapid growth of m_c as θ_{cut} decreases, the exponent ν does not vary substantially as $1/\nu = 0.73 \sim 0.76$ or $\nu = 1.32 \sim 1.37$ over the entire interval $0 < \theta_{cut} < \pi/2$ (see Figs. 2(c) and (d)). These ν values are nearly the same as $\nu = 4/3$ for the lattice percolation [12]. This result physically means that ν hardly depends on θ_{cut} .

Based on Eq. (7) and the data in Figs. 2(a) and (b), we estimate γ/ν as shown in Figs. 2(e) and (f). Neither does the exponent γ/ν vary substantially as $\gamma/\nu = 1.72 \sim 1.73$ or $\gamma = 2.30 \sim 2.35$ over the entire interval $0 < \theta_{cut} < \pi/2$. This value of γ is also very close to $\gamma = 43/18$ for the lattice percolation [12]. The result for γ also physically means that γ hardly depends on θ_{cut} . Figures 2(g) and (h) show that S , with the obtained exponents, satisfies the scaling relation in Eq. (5)

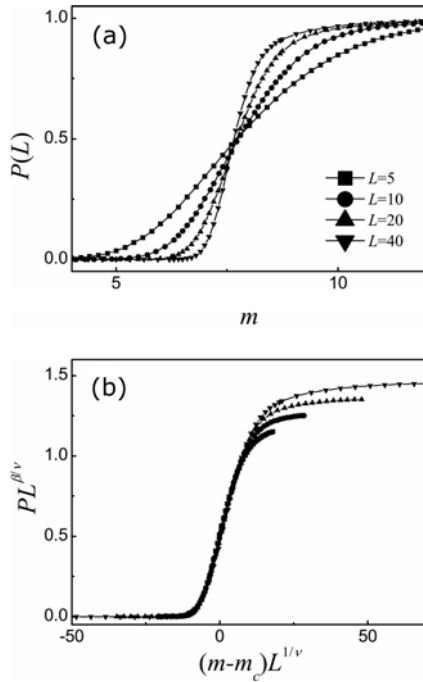


Fig. 4. (a) P against m for various L when $\theta_{cut} = \pi/4$. (b) The data in (a) collapse to the scaling function in Eq. (8).

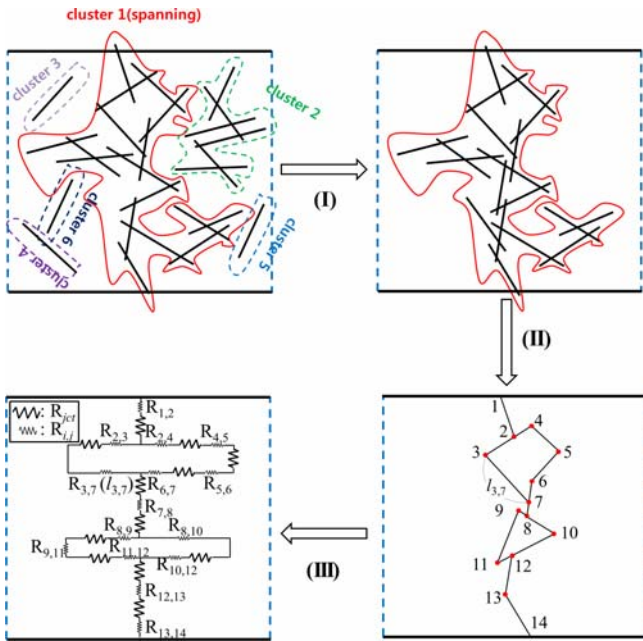


Fig. 5. (Color online) Composition of the random resistor network from a configuration of sticks. Step I: Identification of the spanning cluster. Step II: Construction of the backbone of the cluster. Step III: Resistance as $R_{ij} = \ell_{ij}$ is assigned to the part of the stick between the junctions i and j . Junction resistance R_{jct} is assigned to each junction.

regardless of θ_{cut} .

Figure 4(a) shows the order parameter P of the percolation transition. The order parameter $P(m, L)$ is de-

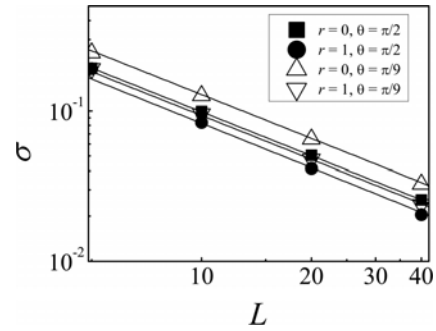


Fig. 6. Conductivity σ against $L (= \ell^{-1})$ for various ratios $r = R_{jct}/R_{NT}$ for (a) $\theta_{cut} = \pi/2$ and (b) $\theta_{cut} = \pi/9$. Each line is the fit of σ to Eq. (9) with a common exponent $t/\nu \simeq 0.96$.

defined as $P(m, L) = N_p/N$, where N_p is the number of sticks in the spanning cluster. $P(m, L)$ for $m \simeq m_c$ or the critical region [12] satisfies the FSS ansatz

$$P(m, L) = L^{-\beta/\nu} g((m - m_c)L^{1/\nu}), \quad (8)$$

as shown in Fig. 4(b). Here, the scaling function $g(x)$ scales as $g(x) \sim x^\beta$ for $x \gg 1$ and as $g(x) = const.$ for $x \ll 1$. In Fig. 4(b), we display the scaling collapse of the obtained P to Eq. (8). For the collapse, we use ν and $m_c(\pi/4)$ in Fig. 2(g) and $\beta = 5/36$ for the lattice percolation transition. Even though only $P(m, L)$ for $\theta_{cut} = \pi/4$ is displayed in Fig. 4 for the sake of simplicity, we confirm that $P(m, L)$ satisfies nearly the same scaling relation (8), regardless of θ_{cut} , with $m_c(\theta_{cut})$ in Fig. 3. This result also supports the fact that the percolation transitions belong to the same universality class as that of the lattice percolation transition regardless of θ_{cut} . The FSSA of the stick percolation systems concludes that the universality class of the percolation transition never changes as the θ_{cut} or the anisotropy of stick alignments varies, even though m_c grows rapidly as θ_{cut} decreases in the interval $\theta_{cut} < 5\pi/18$.

IV. CONDUCTIVITY OF RANDOM RESISTOR PERCOLATION NETWORKS

Based on the results in Sec. III, we now analyze the conductivity of random resistor networks composed from the spanning cluster at the percolation threshold density m_c . For $m > m_c$ or the stick density m above the percolation threshold density m_c , the spanning cluster or the largest cluster is topologically nearly equal to the homogeneous two-dimensional structure. Therefore, the conductivity σ of the resistor network for $m > m_c$ never satisfies a nontrivial power-law behavior like Eq. (1). For $m < m_c$, a spanning cluster does not exist, and the stick network never forms a conducting medium. Thus, a nontrivial power-law like Eq. (1) only occurs on stick networks for $m \simeq m_c$. This is the physical reason we are interested in the conductivity of random resistor networks

composed from the spanning cluster at the percolation threshold density m_c [4,12].

The procedure to compose a random resistor network from a percolation cluster is schematically shown in Fig. 5. To compose the resistor network from the spanning cluster, we took the following three steps: The first step (Step I) is to identify the spanning cluster that connects the top side of $y = L$ to the bottom side of $y = 0$ (See step I in Fig. 5). Since currents only flow through the backbone of the cluster as in the lattice percolation [12], the backbone of the cluster must be identified. The next step (Step II) is to make the backbone by cutting out the dangling sticks or dangling parts of sticks through which no currents flow (See step II in Fig. 5). The third step (Step III) is to assign resistances to the sticks (nanotubes) and to the junctions as in step III of Fig. 5. Here, the stick resistance R_{ij} between the junctions i and j is set to a real length ℓ_{ij} of the part of the stick between the junctions i and j . Because the stick length ℓ is set as a unit in model B and the resistance R_{NT} of a whole stick is one, $R_{ij}(= \ell_{ij}) \leq R_{NT}(= 1)$. R_{jct} is given to each junction.

Balberg *et al.* [9] originally studied the conductivity of the resistor networks for the isotropic alignments of sticks only with R_{jct} . As explained in Sec. I, a recent study [8] argued that the conductivity exponent of the networks never depends on the ratio $r = R_{jct}/R_{NT}$ for the isotropic alignments. In contrast a recent experimental study [4] found a nontrivial behavior as Eq. (1), where the ratio r is experimentally in the interval $7 < r < 70$ [4]. Here, we study the dependence of the conductivity of the networks on the anisotropic alignments or θ_{cut} and on the ratio r .

We use conventional methods based on the Kirchhoff's circuit rule to evaluate the conductivity of the networks [9, 12]. The conductivity σ of the networks at m_c for each set of L , θ_{cut} , and r is obtained by averaging over $10^5 \sim 10^6$ stick configurations. The numerical results for σ are shown in Fig. 6. The conductivity σ at the percolation threshold on a 2D lattice for $r = 0$ is well known to satisfy

$$\sigma \sim L^{-t/\nu}, \quad (9)$$

with $t/\nu \simeq 0.970$ [12,16]. As shown in Fig. 6, the conductivity of isotropic stick networks for $r = 0$ satisfies Eq. (9) very well with $t/\nu = 0.96(1)$, which is very close to the value for the lattice percolation and is nearly identical to the recent result [8] for the stick percolation system. This means that a dynamical property like the conductivity as well as the critical properties of the transition of the stick percolation system, as shown in Sec. III, is the same as that of the lattice percolation system. Furthermore, the result $t/\nu = 0.96(1)$ for $r = 0$ is also reproduced for arbitrary θ_{cut} , as shown in Fig. 6. This also means that the scaling relation (9) for σ should be independent of θ_{cut} or anisotropic alignments as the scaling relations for the critical properties in Sec. III.

The conductivities for cases with finite r ($= 1$) and arbitrary θ_{cut} satisfy Eq. (9) very well with $t/\nu = 0.96(1)$, as shown in in Fig. 6. The conductivities for cases with $r = \infty$ ($R_{NT} = 0$ and $R_{jct} = 1$) and arbitrary θ_{cut} also satisfy Eq. (9) very well with $t/\nu = 0.96(1)$, as for the isotropic case with $r = \infty$ ($R_{NT} = 0$ and $R_{jct} = 1$) [12, 16]. This result means that the conductivity exponent t/ν hardly changes as θ_{cut} changes. σ also satisfies the FSSA, like S and P :

$$\sigma(m, L) = L^{-t/\nu} h((m - m_c)L^{1/\nu}), \quad (10)$$

where the scaling function $h(x)$ scales as $h(x) \sim x^{-t}$ for $x \gg 1$ and $h(x) = const.$ for $x \ll 1$. Thus, in the infinite-size limit or $L \rightarrow \infty$, σ in the critical region or $|m - m_c| \ll 1$ satisfies the relation

$$\sigma \sim (m - m_c)^{-t}. \quad (11)$$

The conductivity exponent t for the infinite-size analysis, as in Eq. (4), satisfies $t = 1.27$, and t never varies as r varies from 0 to ∞ .

The result for the conductivity analysis of the 2D percolation system with an anisotropy of the stick alignments cannot explain the experimentally-measured FSS behavior [4] with a experimentally given resistance ratio $r = R_{jct}/R_{NT}$. The discrepancy might come from two physical reasons. One is that the 2D stick percolation system is physically too simple to be the exact theoretical model system for real SWNT film networks. The other is the data to calculate the experimentally measured FSS behavior [4] have errors that are too large to get Eq. (1). The data in Ref. 4 actually are too scattered to exclude the result in Fig. 6.

V. SUMMARY AND DISCUSSION

By using a FSSA, we analyze the critical properties of percolation transitions in 2D stick systems with anisotropic alignments. The percolation threshold density m_c changes rapidly as θ_{cut} decreases in the interval $\theta_{cut} < 5\pi/18$ as $m_c \sim \theta_{cut}^{-0.9}$. From the practical point of view, understanding the behavior of $m_c(\theta_{cut})$ itself becomes very crucial when we need to design a nanoscale device with minimum nanotube concentration [17]. In contrast, the critical exponents β , γ , and ν barely change as θ_{cut} changes and are nearly the same as those for the lattice percolation transition. This result physically means that the universality of the percolation transition of 2D stick systems is the same as that of the lattice transition regardless of a rapid change of $m_c(\theta_{cut})$.

For the comparison of the experimental result on the conductivity of networks of SWNT bundles, the conductivity of the random resistor networks composed from percolation clusters of stick systems near the percolation transition is calculated. As shown in Fig. 6, the result for the conductivity is nearly independent of the

anisotropic stick alignment or θ_{cut} and the resistance ratio $r = R_{jct}/R_{NT}$, even though the percolation threshold density m_c rapidly changes for small θ_{cut} . This result physically means either that the percolation model of the 2D stick system is physically too simple to be the exact theoretical model system for networks of the SWNT film bundles [4] or that the data to calculate the experimentally-measured FSS behavior [4] have errors that are too large to exclude the result in Fig. 6.

In Du *et al.*'s work [5], the dependences of the conductivity and the percolation properties of nanotube/polymer composites film networks on the anisotropic alignment and the concentration of nanotubes were studied by using both experiments and simulation methods similar to ours. They argued that both the critical properties, like P in Fig. 4 and the conductivity of the networks strongly depend on θ_{cut} . But in the simulation the study was not based on a careful FSSA. The naive result of such strong dependences on θ_{cut} might come from the strong dependence of the percolation threshold m_c on θ_{cut} . As shown in Figs. 2, 4, and 6, a careful FSSA of the simulation data concludes that the scalings of the critical properties and the conductivities do not change as θ_{cut} varies.

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