Conductivity exponents in stick percolation

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On the basis of Monte Carlo simulations, the present work systematically investigates how conductivity exponents depend on the ratio of stick-stick junction resistance to stick resistance for two-dimensional stick percolation. Simulation results suggest that the critical conductivity exponent extracted from size-dependent conductivities of systems exactly at the percolation threshold is independent of the resistance ratio and has a constant value of 1.280 ± 0.014. In contrast, the apparent conductivity exponent extracted from density-dependent conductivities of systems well above the percolation threshold monotonically varies with the resistance ratio, following an error function, and lies in the vicinity of the critical exponent.

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Stick percolation, considered for the first time by Pike and Seager in 1974 [1], has been extensively studied theoretically as an important representative of continuum percolations [2–7]. Recent demonstrations of thin films of networked conducting rodlike nanoparticles, such as silicon nanowires [8] and carbon nanotubes [9], with promising applications in electronics, optoelectronics, and sensors [10–13], have motivated an increasing interest in stick percolation. On one hand, stick percolation has been employed aiming at unveiling the underlying physics of the device performance [14,15] and exploiting new device structures to improve the performance [16]. On the other hand, the rodlike nanoparticles are also in turn used as a model system for study of stick percolation from the experimental viewpoint [17].

Despite the rising interests in stick systems, some critical behaviors in stick percolation have not yet been well understood and remain to be explored. An example concerns controversial conclusions found in the literature for the dependence of conductivity exponent t on the ratio of stick-stick junction resistance (R_j) to stick resistance (R_s), i.e., R_j/R_s. Balberg *et al.* [2] derived for the first time the conductivity exponent for two-dimensional (2D) stick systems as $t = 1.24 \pm 0.03$ in agreement with the universal value $t_0 \approx 1.30$ for lattice percolation [18]. In addition, calculations that assigned only one resistivity value to the sticks or one resistance value to the junctions were found to yield consistent values for t within "experimental uncertainty." [2] On the contrary, by studying the conductivity of carbon nanotube networks, Hecht *et al.* [19] proposed that t for a stick percolation system should vary with R_j/R_s from 0 to the universal value t_0; t approaches t_0 when $R_j \gg R_s$ and t is equal to 0 when $R_j \ll R_s$. The latter was argued to hold by considering the following. When $R_j \ll R_s$, cutting each stick in half would decrease the stick length but would not change the system conductance since R_j is negligible. The conclusion of this oversimplified theoretical treatment is that the system conductance is independent of the stick length and hence t should be 0. Apparently, how t would depend on the resistance ratio R_j/R_s remains to be confirmed. In the present work, we will clarify this dependency based on precise results of extensive and systematic Monte Carlo simulations.

There are usually two schemes for studies of t in a percolation system. The first method evaluates the size dependence of conductivity. According to the finite-size scaling law, the average conductivity $\langle \sigma \rangle$ is expected to vary with the system size L as [20,21]

$$\langle \sigma \rangle = b^{-t/v}[c_1 + c_2 f_2(b) + \cdots], \tag{1}$$

when the stick number density N is equal to the critical density N_c. Here, $b \equiv L/l$, with l being the stick length, c_1 and c_2 are constants, v is the correlation-length exponent, and $f_2(b)$ is the leading correction function with $f_2(b) \to 0$ for $b \to \infty$. This method is favored by theorists and is expected to provide a reliable value for critical exponent t.

The second method relies on the density dependence of conductivity. When N approaches N_c, the conductivity σ of a stick system varies as

$$\sigma \propto (N - N_c)^{\tilde{t}}. \tag{2}$$

Note that Eq. (2) is true only when $L \gg \xi$, where $\xi \propto |N - N_c|^{-v}$ is the correlation length [18]. As N approaches N_c in a finite-size system, there are always finite-sample errors even for very large systems due to the divergence of ξ [2,21]. Rigorously speaking, it is problematic to employ Eq. (2) for extraction of an appropriate value of t for finite-size systems. Nevertheless, it is applicable for extraction of the apparent conductivity exponent $\tilde{t}$ by collecting data at N above, but not very close to, N_c. If N is still within the critical region, $\tilde{t}$ is an approximation of t [2]. When N is well above N_c, though not critical, $\tilde{t}$ can be of particular interest for materials science and electronics involving carbon nanotube networks and nanotube-polymer composites [13,22,23].

It is worth noting that in the previous literature concerning stick percolation, t and $\tilde{t}$ are not carefully distinguished from each other, since few works have employed Eq. (1) to study conductivity exponent. The present work investigates the R_j/R_s dependence for both t and $\tilde{t}$. Two-dimensional widthless isotropic stick percolation is considered on square systems with free boundary conditions. The free boundary conditions are specified in this work just because such stick

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systems are more consistent with the finite-size rodlike nanoparticle networks in practice. Each stick has a fixed length $l=1$ and is centered on a specific site with a given orientation. The system percolates when its left and right boundaries are connected by at least one continuous path consisting of intersecting sticks. The main task is to calculate the conductivity of the percolating system at the critical density N_c with different system sizes L or at different stick density N with the same system size L. The simulations are carried out as follows. First, n $(n=NL^2)$ sticks are produced through randomly generating n uniformly distributed sites (x,y) on the square system for their centers and n random angles θ with respect to the horizontal direction for their orientations. Note that $0\leq x\leq L$, $0\leq y\leq L$, and $0\leq\theta<\pi$. Second, the connectivity property of the sticks is determined. When two sticks intersect at a position between the left and right boundaries of the system, an internal junction is created at the intersection. When a stick intersects with the left or right system boundary, an external junction is created at the intersection. Third, the percolation status is checked for the system. If the system is percolating, all the junctions along the percolation path(s) are marked as "active" junctions. Last, Kirchhoff's current law (KCL) is applied at every active internal junction in order to calculate the system conductivity σ. All sticks have a fixed resistivity ρ. All the internal junctions have a fixed resistance R_j whereas the resistance for all the external junctions is 0. The electric potentials at all active external junctions connecting the left (right) system boundary are set to $V=0$ $(V=1)$. Each active internal junction is associated with two electric potentials, V_i and V_i', corresponding to the electric potentials of the two sticks at the junction position. Since the internal (stick-stick) junctions are of a finite conductance, V_i is usually not equal to V_i'. Then, the equations in terms of KCL are written for both V_i and V_i' at every internal active junction. By solving these equations, σ can be readily represented by the sum of currents flowing into (out of) all external junctions connecting the right (left) boundary. In order to exclude the effect of statistical uncertainties and investigate the intrinsic dependence of conductivity exponents on R_j/R_s $(R_s=\rho l)$, for each simulation realization, the internal junctions are associated with a series of different resistances R_j and the system conductivity σ is calculated for each R_j. Therefore, one can obtain σ of exactly the same stick system but with different resistance ratio R_j/R_s.

Several optimizations have been performed in this work in order to improve the simulation efficiency. During the determination of the connectivity property of the sticks, each stick is first registered into a subcell (a square of unity length) in which its center lies. It is only necessary to check the connectivity property between a chosen stick and any of those belonging to the same subcell or the neighboring subcells [24]. Before applying KCL to establish systems of linear equations for the active internal junctions, all dangling sticks along with the corresponding junctions that do not carry any current [18] are erased. Finally, since the matrices corresponding to the established systems of linear equations are sparse, symmetric, and positive definite, the preconditioned conjugate gradient method (with Jacobi preconditioner) [25,26] is implemented to solve such large systems of linear equations in an iterative way. These optimizations enable us to perform simulations of relatively large systems for stick percolation.

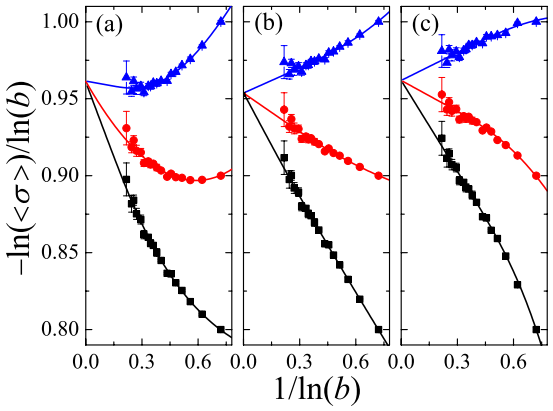

FIG. 1. (Color online) Normalized arithmetic means $\langle\sigma\rangle$ (symbols) and standard deviations (error bars) of the batch arithmetic (■), geometric (●), and harmonic (▲) mean conductivities of stick systems at $N_c=5.637\ 26$ with b varying from 4 to 100, plotted as $-\ln(\langle\sigma\rangle)/\ln(b)$ against $1/\ln(b)$, for (a) $R_j/R_s=10^{-5}$, (b) $R_j/R_s=1$, and (c) $R_j/R_s=10^5$. The intercepts yield t/υ. Solid curves are robust fittings to Eq. (3) with $f_2(b)$ being either the logarithmic or the power correction. In order to exhibit all the data clearly, the $\langle\sigma\rangle$ have been normalized.

A convenient way to extract t is to rewrite Eq. (1) as

$$-\frac{\ln\langle\sigma\rangle}{\ln b}=\frac{t}{\upsilon}-\frac{\ln[c_1+c_2f_2(b)+\cdots]}{\ln b}. \tag{3}$$

Therefore, plotting $-\ln\langle\sigma\rangle/\ln b$ against $1/\ln b$ can give t/υ as the intercept. Figure 1 shows such plots of some of our simulated conductivity for different R_j/R_s at the critical density $N_c=5.637\ 26$ [24]. Note that the simulated conductivity of stick percolation at N_c varies so much among individual realizations that, even with a large number of realizations, the relative standard deviations for the three kinds of mean (arithmetic, geometric, and harmonic) conductivities are all typically around 30–50 %. This phenomenon is different from that in lattice percolation [20,21] where the relative deviations are usually less than 1%. Such large deviations make the extraction of t/υ lack of physical meaning. However, it is also found that all three mean conductivities are rather stable when the number of realizations is sufficiently large. Therefore, five batches of simulations were performed for every b, each of which consists of over $6\times10^5/b^2$ percolating realizations. In this work, $b=4$, 5, 6, 7, 8, 9, 10, 12, 14, 16, 18, 20, 25, 30, 40, 50, 60, and 100. The final results are represented by arithmetic means and standard deviations for the three different mean conductivities of the five batches, as shown by the symbols and error bars in Fig. 1, respectively. All the relative standard deviations are less than 5% and the mean conductivities are thus expected to be reliable for further data analysis.

In order to extrapolate the data to $b=\infty$ and obtain the intercept t/υ, nonlinear regressions with robust fitting [25,27] have been performed with specific correction functions $f_2(b)$. As usual [20], both the logarithmic correction $f_2(b)=1/\ln b$ and the power correction $f_2(b)=b^{-w}$ are uti-

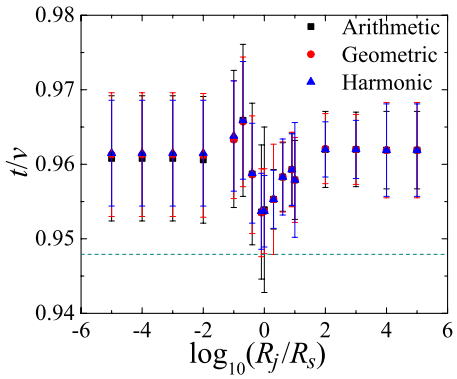

FIG. 2. (Color online) R_j/R_s dependence of t/v extracted from the three kinds of mean conductivities in terms of Eq. (3). Error bars represent the 95% confidence intervals. Horizontal dashed line represents the Alexander-Orbach conjecture.

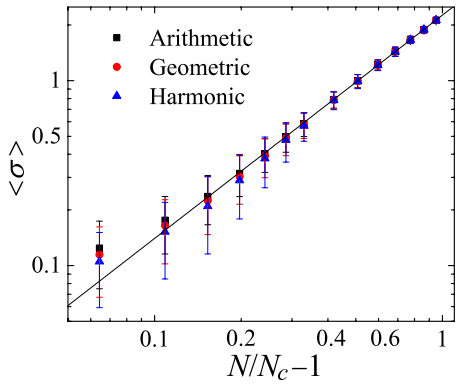

FIG. 3. (Color online) Three mean conductivities as a function of $(N\text{-}N_c)/N_c$ for stick system of $b=20$ and with $R_j/R_s=10^{-5}$. Error bars represent the corresponding standard deviations over all ($\sim$1000) individual realizations. The slope of the fitting line gives $\tilde{t}$.

lized. If only the smallest chi-square (χ^2), the sum of the squared residuals weighted by the reciprocal deviations [25], is preferred, the power correction is superior, with the order w varying from 0.5 to 4. However, the mere pursuit of the smallest χ^2 is not absolutely reasonable since it causes relatively large variations among the extracted t/v from the three mean conductivities and hence also makes it difficult in identifying the true value. Consequently, no preference is put to power correction or logarithmic correction in this work. Both of them are investigated for all three mean conductivities and the optimal set of corrections is determined as those producing the minimum variation among the three extracted t/v and still of comparable χ^2 to the minimum one. The variations are measured by the standard deviation of the three extracted t/v, weighted by the reciprocal of the uncertainties. The solid curves in Fig. 1 represent the fitting results with the optimal sets of corrections. In Fig. 1(a), the optimal set follows this order: power ($w=0.9$), logarithm, and power ($w=1.1$) for arithmetic, geometric, and harmonic mean conductivities, respectively. In Fig. 1(b), it is power ($w=0.7$), power ($w=1.6$), and power ($w=2.4$), and in Fig. 1(c), it becomes power ($w=3.6$), power ($w=2.4$), and power ($w=2.9$).

Figure 2 plots the extracted t/v against the resistance ratio R_j/R_s. It is clearly shown that t/v is independent of R_j/R_s. With R_j/R_s varying from 10^{-5} to 10^5, the variation of t/v is within the experimental uncertainty region. Taking the weighted (by the reciprocal of uncertainties) arithmetic mean of all the measured t/v as the final result, we obtain t in stick percolation as

$$t/v = 0.960 \pm 0.010. \tag{4}$$

Here, the uncertainty is estimated to cover most of the error bars in Fig. 2. This value is consistent with, but slightly lower than, those precise results (around 0.970) reported for lattice percolation [20,28]. As a result of great challenges in calculating conductance for large-size continuum systems, this work considers a few large-size systems. As each of them is of a small number of realizations due to limited computational capacity, relatively large deviations are anticipated. Therefore, the χ^2 fits put less weight to the large-size systems, which can be inferred from Fig. 1 where the data points for the largest b deviate obviously from the fits. This might result in a slight underestimation of t/v in this work. However, most of the t/v values in this work are evidently higher than the Alexander-Orbach conjecture of $t/v=91/96 \approx 0.948$ for 2D percolation [20,28] (shown as a horizontal dashed line in Fig. 2). Hence, this work also invalidates the conjecture from the view of continuum percolation. By taking $v=4/3$, which has actually been implicitly demonstrated in our previous study on finite-size scaling of stick percolation [24], we finally obtain the critical conductivity exponent independent of R_j/R_s as

$$t = 1.280 \pm 0.014. \tag{5}$$

This value is in better agreement with the universal critical exponent t_0.

In order to study the dependence of $\tilde{t}$ on R_j/R_s, conductivities were calculated, as an example, for stick systems of size $b=20$ with different R_j/R_s at N well above N_c. Figure 3 shows the simulation results for $R_j/R_s=10^{-5}$. Different from those obtained exactly at N_c, the conductivity deviations among individual realizations become smaller with increasing N. The symbols and the error bars in Fig. 3 represent, respectively, the true mean conductivities and standard deviations calculated over $\sim$1000 individual realizations. Through linear robust fitting of Eq. (2) to the data in the log-log plot, $\tilde{t}$ can be obtained as the slope of the fitting line. Figure 4 explicitly displays the dependence of $\tilde{t}$ on R_j/R_s. It becomes immediately clear that $\tilde{t}$ is larger for R_j-dominant ($R_j/R_s > 10^2$) systems than for R_s-dominant ($R_j/R_s < 10^{-2}$) systems. This difference in $\tilde{t}$ is considered reliable since it has already gone far beyond the uncertainty region. In addition, $\tilde{t}$ monotonically increases with increasing R_j/R_s throughout the range of R_j/R_s and can be well described by the error function below

$$\tilde{t}(x) = \tilde{t}_0 + A\ \mathrm{erf}(x), \tag{6}$$

where $x=\log_{10}(R_j/R_s)$, $\tilde{t}_0$ and A are constants, and $\mathrm{erf}(x)$ is the error function. The robust nonlinear fitting of all the data in Fig. 4 gives $\tilde{t}_0=1.314\pm0.002$ and $A=0.108\pm0.003$. Note

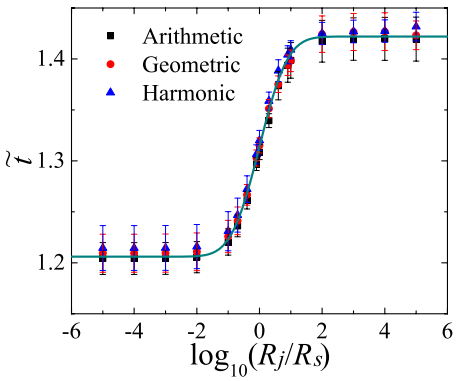

FIG. 4. (Color online) R_j/R_s dependence of $\tilde{t}$ extracted, in terms of Eq. (2), from the three kinds of mean conductivities for stick system of $b=20$ and with $(N-N_c)/N_c$ between 0.3 and 1. Error bars represent the 95% confidence intervals. The curve is the robust fitting to Eq. (6).

that $\tilde{t}_0$ and A vary with the density region. In this work, we are only interested in the density region of $0.3<(N\text{-}N_c)/N_c<1$, since only when $(N\text{-}N_c)/N_c>0.3$, the extracted $\tilde{t}$ from the three kinds of mean conductivities agree well with one another, while on the other hand, too high a density tends to pull the systems far away from the critical region. The inclusion of the lower-density region causes poor consistency among the three extracted $\tilde{t}$. However, if N is not too low, each of the extracted $\tilde{t}$ still follows the error-function dependence as expressed by Eq. (6).

It has now become apparent that the R_j/R_s nondependency found by Balberg *et al.* concerns the critical exponent t while the R_j/R_s dependency proposed by Hecht *et al.* is valid for the apparent exponent $\tilde{t}$. Qualitatively, Eq. (6) coincides with the proposal of Hecht *et al.* in that $\tilde{t}$ decreases with decreasing R_j/R_s. However, this dependency expressed by Eq. (6) has a different interpretation. When a stick system is well above the percolation threshold, its percolating clusters have already been well established. In this case, adding a new stick to the system will not significantly change the cluster structure. But if the sticks are all very conductive and the system conductance is governed by the junction resistance (i.e., $R_j \gg R_s$), adding a stick can shortcut many junctions in a cluster and thus significantly increase the system conductance. As such, the system conductance can be very sensitive to the stick density, that is to say, $\tilde{t}$ is large. As revealed in Fig. 4, $\tilde{t}$ can even be greater than t_0. On the contrary, if the sticks are all poorly conducting so that the system conductance is dictated by the high stick resistance (i.e., $R_j \ll R_s$), adding more sticks only leads to negligible conductance changes to the system and hence low values of $\tilde{t}$ are expected. Similarly, the R_j/R_s nondependency of t can be readily understood. Exactly at the percolation threshold, the percolating cluster of a system is critical. Adding a stick, no matter whether of high conductance or of high resistance, tends to impact the cluster backbone and therefore lead to significant conductance changes of the system. As a result, t is insensitive to R_j/R_s.

Furthermore, our results do not quantitatively support Hecht's assumption of $0\leq\tilde{t}\leq t_0$. As shown in Fig. 4, all $\tilde{t}$ are still around the critical exponent t. The upper limit of $\tilde{t}$ can be definitely greater than t_0, while there is no indication for $\tilde{t}$ to approach 0 when $R_j \ll R_s$. A question here is then how one should correctly address the oversimplified theoretical treatment by Hecht *et al.* leading to $\tilde{t}=0$ for $R_j \ll R_s$. Cutting a stick into two without permitting the latter to reallocate or reorient relative to each other imposes implicit constraints to the stick system in consideration. Such a stick system is correlated and the assumptions for isotropic stick systems no longer apply. In isotropic systems, the two half sticks should indeed be allowed to change their locations and orientations in a random fashion.

Finally, it is also worth pointing out that a major reason for the previously reported [13,23] large deviation between the experimentally extracted conductivity exponents and the critical one is likely due to data acquisition in the stick density region far above the percolation threshold. Consequently, the extracted exponents should correspond more to the apparent exponent than to the critical exponent.

In summary, Monte Carlo simulations with proper optimizations have been performed to study the conductivity of 2D isotropic stick percolation systems with size up to 100. The dependence of conductivity exponents on the junction-to-stick resistance ratio R_j/R_s is systematically investigated. Simulation results indicate that the critical conductivity exponents extracted from size-dependent conductivities of systems exactly at the percolation threshold are independent of R_j/R_s. By varying R_j/R_s in a wide region from 10^{-5} to 10^5, the critical exponents are all found at 1.280 ± 0.014. However, the apparent conductivity exponents extracted from density-dependent conductivities of systems well above the percolation threshold are significantly dependent on R_j/R_s. This dependency can be well described with an error function of the logarithm of R_j/R_s. In the investigated density region of the present work, these apparent conductivity exponents vary within a narrow band near the critical conductivity exponent, disagreeing with previously reported assumptions. These results are considered valuable for theoreticians working on continuum percolation and useful for experimentalists interested in materials of rodlike nanoparticles as well as the electronic and optoelectronic devices based on such materials.

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[1] G. H. Pike and C. H. Seager, Phys. Rev. B **10**, 1421 (1974).
[2] I. Balberg, N. Binenbaum, and C. H. Anderson, Phys. Rev. Lett. **51**, 1605 (1983).
[3] I. Balberg and N. Binenbaum, Phys. Rev. B **28**, 3799 (1983).
[4] I. Balberg, N. Binenbaum, and N. Wagner, Phys. Rev. Lett. **52**, 1465 (1984).
[5] I. Balberg, C. H. Anderson, S. Alexander, and N. Wagner, Phys. Rev. B **30**, 3933 (1984).
[6] W. J. Boudville and T. C. McGill, Phys. Rev. B **39**, 369 (1989).
[7] Z. Néda, R. Florian, and Y. Brechet, Phys. Rev. E **59**, 3717 (1999).
[8] X. Duan, C. Niu, V. Sahi, J. Chen, J. W. Parce, S. Empedocles, and J. L. Goldman, Nature (London) **425**, 274 (2003).
[9] E. S. Snow, J. P. Novak, P. M. Campbell, and D. Park, Appl. Phys. Lett. **82**, 2145 (2003).
[10] Q. Cao, H.-S. Kim, N. Pimparkar, J. P. Kulkarni, C. Wang, M. Shim, K. Roy, M. A. Alam, and J. A. Rogers, Nature (London) **454**, 495 (2008).
[11] Q. Cao and J. A. Rogers, Nano Res. **1**, 259 (2008); Adv. Mater. **21**, 29 (2009).
[12] R. A. Street, Adv. Mater. **21**, 2007 (2009).
[13] L. Hu, D. S. Hecht, and G. Grüner, Nano Lett. **4**, 2513 (2004).
[14] S. Kumar, J. Y. Murthy, and M. A. Alam, Phys. Rev. Lett. **95**, 066802 (2005).
[15] J. Li and S.-L. Zhang, Phys. Rev. B **79**, 155434 (2009).
[16] J. Li, Z.-B. Zhang, M. Östling, and S.-L. Zhang, Appl. Phys. Lett. **92**, 133103 (2008).
[17] B. Vigolo, C. Coulon, M. Maugey, C. Zakri, and P. Poulin, Science **309**, 920 (2005).
[18] D. Stauffer and A. Aharony, *Introduction to Percolation Theory*, 2nd revised ed. (Taylor and Francis, London, 2003).
[19] D. Hecht, L. Hu, and G. Grüner, Appl. Phys. Lett. **89**, 133112 (2006).
[20] C. J. Lobb and D. J. Frank, Phys. Rev. B **30**, 4090 (1984).
[21] D. B. Gingold and C. J. Lobb, Phys. Rev. B **42**, 8220 (1990).
[22] N. Grossiord, J. Loos, L. van Laake, M. Maugey, C. Zakri, C. E. Koning, and A. J. Hart, Adv. Funct. Mater. **18**, 3226 (2008).
[23] H. E. Unalan, G. Fanchini, A. Kanwal, A. D. Pasquier, and M. Chhowalla, Nano Lett. **6**, 677 (2006).
[24] J. Li and S.-L. Zhang, Phys. Rev. E **80**, 040104(R) (2009).
[25] W. H. Press, S. A. Teukolsky, W. T. Vetterling, and B. P. Flannery, *Numerical Recipes: The Art of Scientific Computing*, 3rd ed. (Cambridge University Press, New York, 2007).
[26] http://en.wikipedia.org/wiki/Conjugate_gradient_method
[27] http://www.mathworks.com/access/helpdesk/help/toolbox/stats/nlinfit.html
[28] J. G. Zabolitzky, Phys. Rev. B **30**, 4077 (1984); H. J. Herrmann, B. Derrida, and J. Vannimenus, *ibid.* **30**, 4080 (1984); D. C. Hong, S. Havlin, H. J. Herrmann, and H. E. Stanley, *ibid.* **30**, 4083 (1984).