Resistivity in percolation networks of one-dimensional elements with a length distribution

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One-dimensional (1D) nanoelements, such as nanotubes and nanowires, making up percolation networks are typically modeled as fixed length sticks in order to calculate their electrical properties. In reality, however, the lengths of these 1D nanoelements comprising such networks are not constant, rather they exhibit a length distribution. Using Monte Carlo simulations, we have studied the effect of this nanotube and/or nanowire length distribution on the resistivity in 1D nanoelement percolation networks. We find that, for junction resistance-dominated random networks, the resistivity correlates with root-mean-square element length, whereas for element resistance-dominated random networks, the resistivity scales with average element length. If the elements are preferentially aligned, we find that these two trends shift toward higher power means. We explain the physical origins of these simulation results using geometrical arguments. These results emphasize the importance of the element length distribution in determining the resistivity in these networks.

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There has been significant recent interest in percolation networks made up of one-dimensional (1D) nanoscale objects, such as nanotubes and nanowires, for applications such as thin film transistors [1], flexible microelectronics [2], microelectromechanical systems (MEMS) and chemical sensors [3,4], and transparent, conductive electrodes in photovoltaic and optoelectronic devices [5–7]. Although there exist numerous experimental works on the subject, comparatively few have addressed modeling and simulation of their properties. All of the recent simulation work on the electrical and thermal conductivity of single-walled carbon nanotube (CNT) networks and films have modeled the nanotubes as 1D widthless sticks of a fixed length [8–10]. In particular, a recent simulation study of the effect of element (nanotube) length on the resistivity in nanotube films has shown that the resistivity obeys an inverse power law with element length in the case that all elements are of fixed length [9]. In other words, $\rho \propto l^{-\gamma}$, where ρ is the resistivity, l is the element length, and γ is the critical exponent. In reality, however, the lengths of these 1D nanoelements comprising such networks are usually not constant, rather they exhibit a length distribution [11]. Although the effect of fixed length has been studied, the effect of this length distribution on the resistivity in these percolation networks has not been investigated previously.

In this Brief Report, we systematically study the effect of the nanotube and/or nanowire length distribution on the resistivity in 1D nanoelement percolation networks for the two limiting cases of the resistance. We find that, for junction resistance-dominated random networks, such as CNT films [9–12], the resistivity correlates with root-mean-square (rms) element length and not the average length. For element resistance-dominated random networks, on the other hand, the resistivity is found to scale with average element length. If the 1D elements in the network are preferentially aligned, we find that these two trends shift toward higher power means. We explain the physical origins of these simulation results using geometrical arguments. These results not only provide fundamental insights into the physics of percolation transport in 1D nanoelement networks, but also provide guidance for future experimental work by pointing out the relevant parameters with which the resistivity correlates.

The study of the electrical characteristics of 1D nanoelement networks followed a Monte Carlo approach, as described previously [9,10]. The simulated three-dimensional networks are comprised of an array of stacked two-dimensional (2D) planes of randomly dispersed, stick-like elements [see Fig. 1(a)] between the source and drain electrodes, where each plane represents a small range of vertical distances from the substrate. Constraining each element to one plane models experimental evidence that carbon nanotubes in films orient themselves mostly parallel to the sub-

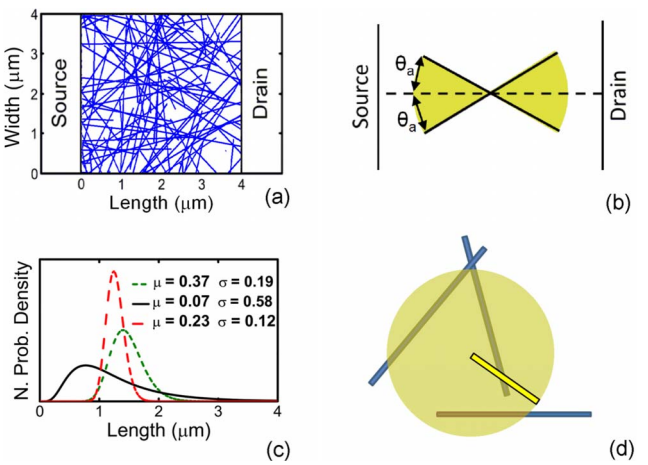

FIG. 1. (Color online) (a) A random 2D nanoelement network generated using a Monte Carlo process, showing the source and drain electrodes. (b) Illustration of the definition of the alignment angle θ_a. (c) Various lognormal length distributions with the solid and long-dashed curves sharing the same average length and solid and short-dashed curves sharing the same rms length. (d) Illustration of the circular probabilistic self-area swept out by a particular element (shaded).

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strate [12]. Each data point in this paper is the statistical average of 300 such randomly generated meshes. The simulation first places each element randomly in its layer with a length l, conforming to a length distribution $\Psi(l)$, and a random orientation θ. For aligned networks, which are also studied in this paper, θ is limited to the range $-\theta_a \leqslant \theta \leqslant \theta_a$ and $180-\theta_a \leqslant \theta \leqslant 180+\theta_a$, where θ_a is defined as the alignment angle, as shown in Fig. 1(b) [10]. For this study, we employ the lognormal distribution which is given by

$$\Psi(l) = \frac{1}{l\sigma\sqrt{2\pi}} \exp\left(-\frac{[\ln(l)-\mu]^2}{2\sigma^2}\right).$$

Figure 1(c) shows sample lognormal distributions with different σ and μ values, where σ and μ are the standard deviation and the mean, respectively.

The locations of all junctions are found by observing intersections between elements on the same layer and between nearest-neighbor layers under the assumptions that each layer is thick enough to allow same-layer interactions yet thin enough for nearest-layer interactions to be equally likely and that the networks are sparse enough to avoid the effects of geometric constraints on the amount of junctions that can occur on a particular element. Kirchhoff's current law is imposed at each junction to calculate the resistivity of the network by solving a set of nodal equations [9,10].

Two sources of resistance contribute to the overall resistivity of a network: Element-element junction resistance and the lengthwise resistance of each element itself. As a result, junctions are modeled by an effective resistance R_J at each junction and the lengthwise resistance along each element between two junctions a distance d apart is calculated by $R_d = R_0 d/\lambda$, where λ is the mean free path, and R_0 is a constant, which corresponds to the resistance of a 1D element having a length equal to the mean free path. By these definitions, there are two limiting cases for the resistivity of the network: (1) $R_J \gg R_0$, which is the junction-dominated case and (2) $R_J \ll R_0$, which is the element-dominated case. For example, in the case of CNT films, junction resistance has been found to dominate [9,11,13], but in general this may not be true; element resistance could dominate for nanoelements such as large diameter semiconducting nanowires.

Figure 2(a) shows normalized resistivity versus average 1D element length given by $\langle l \rangle = \exp(\sigma^2/2+\mu)$ for an unaligned, junction-dominated network for four different σ-μ relationships represented by the four data series. The standard error bars are not larger than the size of the symbols for all figures. Strong scaling with length, which has been reported before [9], is apparent for each data series. However, the resistivities for different length distributions at a fixed average length are distinctly separate, suggesting that, contrary to what one might initially expect [9,11], the network resistivity is not an explicit function of average element length in this case.

As a result of the monotonic decreasing nature of resistivity in Fig. 2(a), and noting that the widest element length distributions produce the least resistivity, we plot in Fig. 2(b) the resistivity versus rms (second power mean) element length, given by $\sqrt{\langle l^2 \rangle} = \sqrt{\int_{-\infty}^{\infty} l^2 \Psi(l) dl} = \exp(\mu+\sigma^2)$, for four different σ-μ relationships. Because the rms value of any distribution is always greater than or equal to its mean, the effect of Fig. 2(b) is to shift the data series corresponding to wider distributions in Fig. 2(a) to the right so that they coincide with each other. This shows that the rms length, and not average length, is the relevant length parameter determining network resistivity.

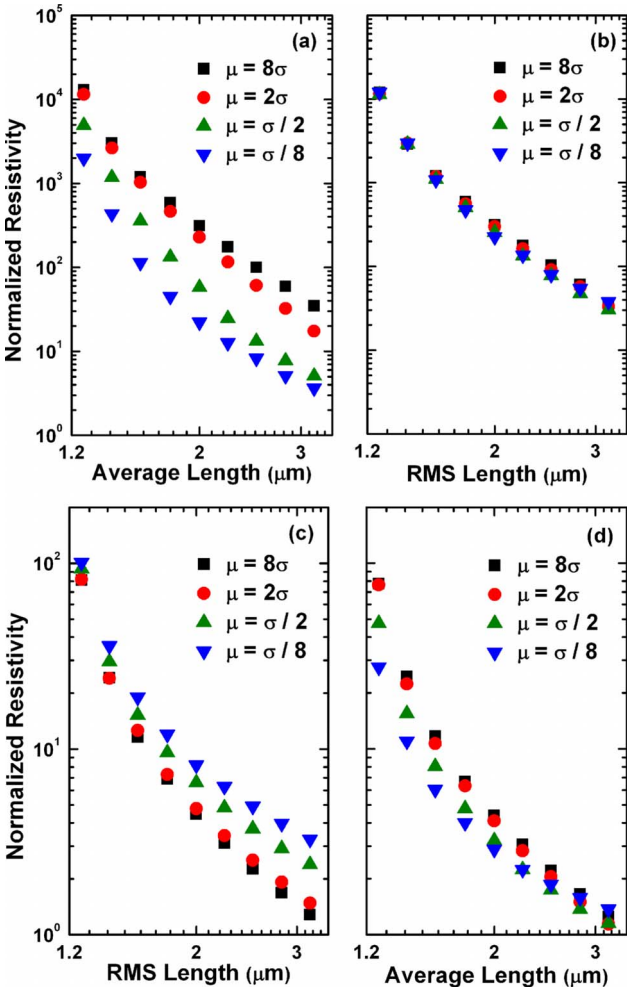

FIG. 2. (Color online) Log-log plot of normalized resistivity of a junction-dominated network versus (a) average element length and (b) rms length for four different σ-μ relationships (labeled by different symbols). The network has a thickness of five layers with device length $L=15$ μm, device width $W=15$ μm, and density per layer $n=1.2$ μm^{-2}. Log-log plot of normalized resistivity of a element-dominated network versus (c) rms length and (d) average length using the same device parameters as in parts (a) and (b).

Figure 1(d) rationalizes the result of Fig. 2(b) by illustrating that within each network layer, each 1D element sweeps out a circular probabilistic "self-area" that is the superposition of all orientations it may take once anchored to a point. This self-area represents all possible points of contact with other nanostuctures, and as a result, the larger this area, the more junctions that this element can make, and the lower the resistivity of a junction-dominated network. Since the concept of rms length, by weighing the length distribution by l^2, is equivalent to averaging of areas swept out by an element of length l, the resistivity correlates with rms length for a

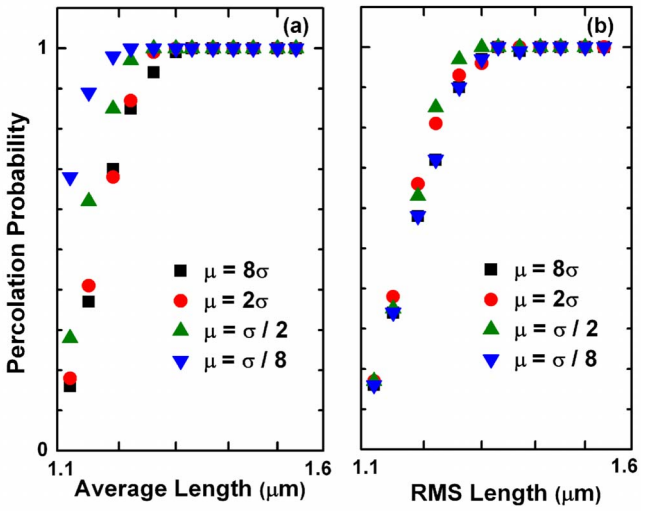

FIG. 3. (Color online) Percolation probability versus log (a) average and (b) rms length for the resistivity data in Fig. 2. Only two plots are shown since percolation probability is a geometrical quantity which is independent of whether a network is junction or element dominated.

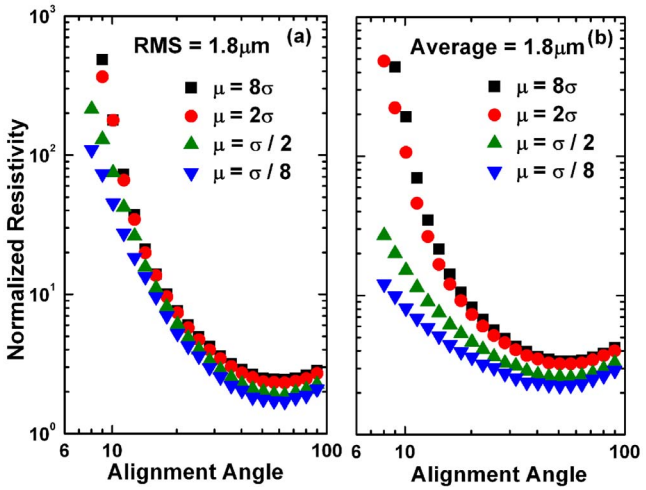

FIG. 4. (Color online) Log-log plot of normalized resistivity versus alignment angle for (a) junction-dominated network with fixed rms length=1.8 μm and (b) element-dominated network with fixed average length=1.8 μm for four different σ-μ relationships, as labeled by different symbols. Both plots show a progressive divergence of the data for highly aligned networks.

junction-dominated network. It can further be shown that this rms correlation is independent of the functional form of the element length distribution [14]. For a network with fixed element length, the rms length is equal to the average length, and as a result, the effect of one length metric is indistinguishable from the other [9].

For the element-dominated case, however, the situation is quite different. Figure 2(c) shows normalized resistivity versus rms length for a element-dominated network having the same dimensions as in Fig. 2(a) for four different σ-μ relationships. The four series separate, suggesting that, in contrast to the junction-dominated case, the network resistivity is not an explicit function of rms length for element-dominated networks. Figure 2(d), on the other hand, shows normalized resistivity versus average length for four different σ-μ relationships. It is evident that for large average lengths, all four length distributions show convergence to a singular point for each average length. This suggests a good correlation between resistivity and average length for element-dominated networks. In this case, the network resistivity is independent of the number of junctions, but depends on the length and number of conducting paths between the source and drain electrodes, which correlate with average length.

However, it is evident from Fig. 2(d) that as the average length decreases, the four data series increasingly diverge. Furthermore, Fig. 2(c) shows a corresponding convergence in the resistivity versus rms length plots for small lengths. These observations can be better understood by plotting the percolation probability (defined as the probability of finding at least one conducting path between the source and drain) versus average length and rms length, as shown in Figs. 3(a) and 3(b), respectively. Percolation probability is a geometrical quantity which is independent of whether a network is junction or element dominated. It is clear from Fig. 3 that when the percolation probability drops below 1, it shows a better correlation with rms length compared to average length. Physically, this is due to the fact that a conducting path may only extend to another element at a junction, which is proportional to the circular probabilistic self-area swept out by each 1D element, as previously shown in Fig. 1(d) and discussed in previous work [15,16]. Since percolation probability correlates with rms length, which is itself a formulation of average self-area, percolation effects are seen to drive the resistivity scaling toward rms length for short element lengths, as observed in Figs. 2(c) and 2(d).

At this point, it is worth discussing the applicability of the concept of "excluded area" introduced by Balberg *et al.* [16] in determining the percolation threshold [17] for a 2D network of elements with a length distribution. A logical extension of the excluded area idea for a system of widthless sticks with a length distibution $\Psi(l)$ gives the average excluded area A as $\langle A\rangle=\langle l\rangle^2\langle\sin|\theta_i-\theta_j|\rangle$, where θ_i and θ_j are the angles two interacting sticks labeled by i and j make with the horizontal axis, respectively (see Fig. 1 in Ref. [16]). This result, which is proportional to average length, contradicts the simulation results in Fig. 3 and other Monte Carlo simulations reported in the literature. As also discussed by Balberg *et al.* [16], the proper averaging of the excluded area suggested by our Monte Carlo simulations is $\langle A\rangle=\langle l^2\rangle\langle\sin|\theta_i-\theta_j|\rangle$, which is proportional to rms length. This implies that the concept of self-area associated with an element illustrated in Fig. 1(d) explains the simulation results near the percolation threshold better when there is a length distribution, although its geometrical meaning is not as transparent as that of excluded area.

Finally, we have also studied the effect of alignment of elements in the network on the resistivity for different length distributions. Figure 4(a) shows normalized resistivity as a function of alignment angle θ_a for the junction-dominated case, where each data series has a fixed rms of 1.8 μm, but a different σ-μ relationship. As reported previously for the

case of fixed element length [10,18], the curves are not monotonic, but rather they reach a minimum at some intermediate θ_a that is neither perfectly aligned nor unaligned for all cases of σ and μ.

As the alignment angle decreases and imposes tighter constraints on the orientations of the elements, the rms correlation observed for junction-dominated random networks vanishes. A similar result is observed for the average length correlation of element-dominated networks in Fig. 4(b), where each data series has a fixed average length of 1.8 μm, but a different σ-μ relationship. Similar to the explanation of Fig. 2(b), since wider length distributions exhibit a smaller resistivity in Fig. 4 and $\sqrt[n]{\langle l^n \rangle} \geq \sqrt[m]{\langle l^m \rangle}$ for $n > m$ for a particular length distribution, these results indicate that the correlation for aligned networks shifts towards higher power mean lengths, namely towards the third power mean $\sqrt[3]{\langle l^3 \rangle}$ for both resistive regimes. Indeed, the increasing divergence of the plots with alignment represents an increasing reliance on longer elements to carry charge from source to drain.

In an attempt to explain this result, first let us consider a network with fixed element length l [16,18,19]. For aligned elements (i.e., small θ_a), we compute the average longitudinal (source-drain direction) displacement of an element for all θ, given by $2l\int_{-\theta_a}^{\theta_a} \psi_\theta \cos(\theta) d\theta = l\frac{\sin(\theta_a)}{\theta_a}$, where $\psi_\theta = (4\theta_a)^{-1}$ is the uniform angular distribution. Physically, this quantity measures how much a particular conducting element aids in ferrying charge from source to drain, which we see increases with decreasing θ_a. This effect competes with the corresponding shrinking probabilistic contact self-area, which rationalizes the region of minimum resistivity observed in Fig. 4 for slight network anisotropy as the region where the two effects reverse in dominance. In order to combine the contribution of these effects into one quantity, a natural choice is to multiply these effects together; interestingly, the result is a formulation proportional to l^3. Although it seems possible to generalize this to $\langle l^3 \rangle$ (and therefore $\sqrt[3]{\langle l^3 \rangle}$) for a length distribution, the literature suggests there is no obvious correlation for systems with even slight anisotropy, such as alignment [16,20]. Regardless, increasing alignment can be seen to increase the relative weight of longer elements to the overall conduction, hence driving the resistivity scaling toward higher power mean lengths.

In summary, we have performed Monte Carlo simulations of 1D element percolation networks for different length distributions in the both junction- and element-dominated cases to determine how each affected resistivity scaling with length. We have observed that network resistivity correlates well with rms length for junction-dominated and with average length for element-dominated networks. In the latter case, percolation effects drive the correlation towards rms length for short average lengths. Furthermore, in each case, alignment of nanotubes and/or nanowires in the network places increasing weight on the longest elements, shifting the correlation to higher power means. These results not only provide fundamental insights into the physics of the percolation transport in 1D nanoelement networks, but also emphasize the importance of taking the nanoelement length distribution into account when using these networks in potential device applications.

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