Limits of Performance Gain of Aligned CNT Over Randomized Network: Theoretical Predictions and Experimental Validation

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***Abstract*—Nanobundle thin-film transistors (NB-TFTs) that are based on *random* networks of single-walled carbon nanotubes are often regarded as high performance alternative to amorphous-Si technology for various macroelectronic applications involving sensors and displays. Here, we use stick-percolation model to study the effect of collective (stick) alignment on the performance of NB-TFTs. For long-channel TFT, small degree of alignment *improves* the drain current due to the reduction of average path length; however, near-parallel alignment *degrades* the current rapidly, reflecting the decrease in the number of connecting paths bridging the source/drain. In this paper, we 1) use a recently developed alignment technique to fabricate NB-TFT devices with multiple densities D, alignment θ, stick length L_S, and channel length L_C; 2) interpret the experimental data with a stick-percolation model to develop a comprehensive theory of NB-TFT for arbitrary D, θ, L_S, and L_C; and 3) demonstrate theoretically and experimentally the feasibility of fivefold enhancement in current gain with optimized transistor structure.**

***Index Terms*—Aligned carbon nanotube (CNT) networks, percolation threshold, random CNT networks, stick percolation, thin-film transistors (TFTs), transistor models.**

I. Introduction

RECENTLY, THERE has been significant interest in higher performance alternative to amorphous-Si technology for the novel applications in macroelectronic displays, chemical/biological sensors, photovoltaics, flexible electronics, etc. [1]–[3]. Plastic/glass substrates are desired for these applications which require low temperature manufacturing. Single-walled carbon nanotube (SWCNT)-based thin-film transistors (TFTs) have demonstrated excellent mobility μ [1], but they suffer from low drive current I_D and poor yield due to difficulties in precise placement of individual tubes. In contrast, nanobundle TFTs (NB-TFTs) that are based on *random* 2-D networks [4] of carbon nanotubes (CNTs) address the tube-placement issue and provide higher I_D through multiple percolating paths [5]–[7], bridging the source (S) and the drain (D). Obviously, the network must be above the percolation threshold so that at least one uninterrupted path exists between any two points on an infinite network. The current I_D is proportional to the number of parallel percolating paths and scales inversely with the average path length. Partial alignment of the tubes decreases the average path length between S/D (increases I_D) while reducing the number of available parallel paths between S/D (reduces I_D), giving a point of optimal alignment. In this paper, we systematically vary CNT alignment, by experiments and by simulations, from completely random [Fig. 1(e)] to fully aligned [8]–[10] [Fig. 1(a)] network and study the effect of degree of alignment on NB-TFT drain current I_D. We find that I_D of an optimally aligned network is ∼20%–40% higher for a long-channel ($L_C > L_S$) NB-TFT and, at best, ∼100% higher for a short-channel ($L_C < L_S$) NB-TFT as compared to I_D of corresponding random network transistor.

A. Stick-Percolation Model

We developed a finite-size 2-D numerical stick-percolation model for the NB-TFTs by generalizing the random-network theory discussed in [11]. The model [12], [13] randomly populates a 2-D grid by sticks of length L_S and orientation θ with probability density function (pdf) consistent with the experimental conditions (Fig. 1). For each simulation, a simple box filter with different θ_{avg} is used, for example, $\theta_{\mathrm{avg}} = 45^\circ$ implies that the tubes can make any angle from 0° to 90° with equal probability, i.e., the network is completely random. Here, we only consider the first quadrant due to symmetry.

For electrical properties of the film, we presume that one-third of the generated tubes are metallic and the remaining two-third are semiconducting [8]. Since L_C and L_S are much larger than the phonon mean free path, contact resistances are not important and in linear regime ($V_G > V_{\mathrm{TH}}$, $V_D \sim$ small), transport within individual stick segments of this anisotropic stick network is well described by drift-diffusion theory [12], [14], [15]. In the linear regime, small V_D and constant V_G obviate the need to solve the Poisson equation. Further, at low bias, diffusion is negligible and the drift-diffusion equation reduces to $J = q\mu n\, d\varphi/ds$, where n is carrier density, ϕ is the potential, and s is the length along the tube. When combined

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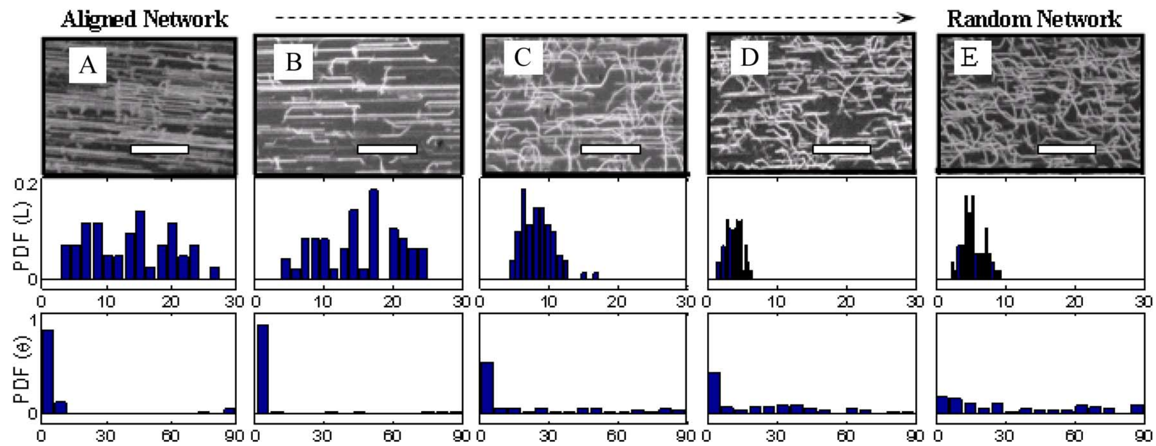

Fig. 1. SEM images of guided growth of SWCNT film on quartz wafers with different alignment achieved by different annealing time: (a) 8 h, (b) 8 h, (c) 60 min, (d) 30 min, and (e) 0 min [8]–[10]. The scale bar is 10 μm. The pdfs of length (in micrometers) and angle distribution are calculated by image analysis of the SEM images. The pdf of angle distribution becomes more uniform while average length reduces with lesser annealing time.

with current continuity equation $dJ/ds = 0$, the potential ϕ_i along tube i is given as

$$\frac{d^2\phi_i}{ds^2} - c_{ij}(\phi_1 - \phi_j) = 0. \qquad (1)$$

Here, $c_{ij} = G_0/G_1$ is the dimensionless charge-transfer coefficient between tubes i and j at their intersection point, G_0 ($\sim$0.1 e^2/h) and G_1 $(qn\mu/\Delta x)$ being the mutual and self conductance of the tubes, respectively [12]. Hundreds of such samples were simulated to accurately reflect the pdf of length and anisotropy distribution in Fig. 1, and the average of these currents is compared to the measured data.

II. Results and Discussion

A. Long-Channel Transistors ($L_C > L_S$)

Fig. 2(a) shows the simulated (red solid curve) and experimental (red circles) results for ON-state current I_D of long-channel NB-TFT versus alignment $\theta_{\rm avg}$. Remarkably, the stick-percolation model quantitatively reproduces three counterintuitive experimental features: 1) I_D is maximized by an optimum theta distribution, which is in between random and aligned networks ($0 < \theta_{\rm avg,opt} < 45°$); 2) I_D of the optimally aligned network is approximately 20%–40% higher compared to I_D of the random NB-TFT; and 3) finally, I_D degrades rapidly as perfect alignment ($\theta_{\rm avg} \rightarrow 0$) is achieved (see Fig. 2(a), solid red curve for $L_C/L_S = 8$. Note that each simulation point reflects average of $\sim$200 statistical samples, and generation of each curve in Fig. 2 requires $\sim$4 h on 50-node Opteron system).

A close analysis of the results shows that the three features of the measured data are actually different manifestations of the same geometrical/physical process. First, note that for $L_C > L_S$, no single tube bridges S/D directly and there is a minimum density D (percolation threshold $D_{\rm perc}$) that allows the onset of tube-to-tube hopping conduction. Furthermore, $D_{\rm perc}$ along the alignment axis increases monotonically with alignment [16]–[19] (i.e., $D_{\rm perc}(\theta_{\rm avg} = 45°) = 4.236^2/\pi L_S^2$ [20] to $D_{\rm perc}(\theta_{\rm avg} \rightarrow 0) \rightarrow \infty$), because increasing alignment reduces the probability of tube crossing. Therefore, although alignment

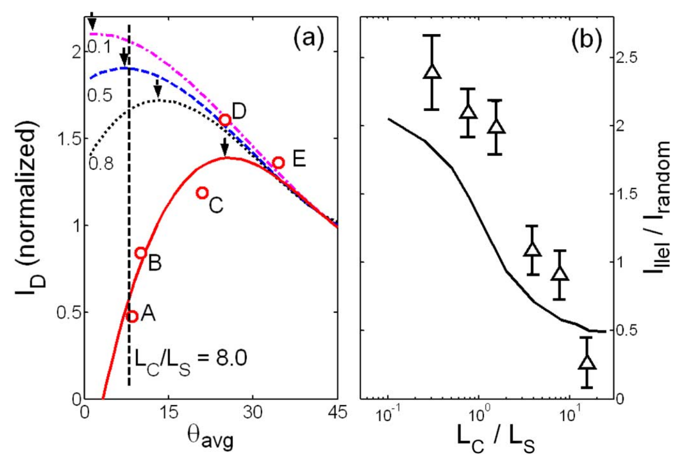

Fig. 2. (a) Simulated ON-state current I_D versus alignment $\theta_{\rm avg}$ for $L_C/L_S = 0.1$, 0.5, 0.8, 8.0. For long-channel ($L_C/L_S = 8 > 1$) NB-TFTs, the symbols (circles) show the experimental results which have three main features: 1) $\theta_{\rm avg,opt}$ gives maximum I_D, for sample D (see Fig. 1 for SEM); 2) $I_{D,\rm opt}/I_{D,\rm random} = 1.2$–1.4; and 3) I_D degrades rapidly as perfect alignment is achieved for samples A and B. For short-channel ($L_C/L_S < 1$) NB-TFTs, the simulations suggest that the optimal alignment angle $\theta_{\rm avg,opt}$ decreases (more alignment needed, see arrows) as L_C/L_S is reduced. (b) (line) Theoretical and (symbol) experimental results for $I_{\|\rm el}/I_{\rm random}$ (corresponding to Fig. 1(a) and (e), respectively) as a function L_C/L_S for $\theta_{\rm avg} \sim 8$ [dotted line in (a)]. For $L_C \ll L_S$, $\theta_{\rm avg,opt} = 0$, i.e., perfectly aligned network in this case is actually optimal. Note that since the network in Fig. 1(e) has a small degree of alignment and is not completely random, the (symbols) experimental results have been normalized accordingly. The error bars show the variations in the experimental results.

reduces the number of sticks required to bridge the S/D, the probability of formation of such bridges (dictated by $D_{\rm perc}$) also reduces with more alignment. Indeed, the quantitative agreement between theory and experiments suggests that for long-channel transistors, the gain through alignment is small and a random network is surprisingly close to being optimal. If, therefore, further current gain over the random network is desired, one must use shorter channel transistors, as discussed in Section II-B.

B. Short-Channel Transistors ($L_C < L_S$)

We use the same numerical stick-percolation model that was used for long-channel transistors, as discussed in

Section II-A, to predict and optimize the electrical performance of short-channel transistors. For $L_C < L_S$, many tubes can bridge source and drain directly ($D_{\rm perc} \rightarrow 0$) for random as well as aligned networks and $I_D \neq 0$ even for $\theta_{\rm avg} \rightarrow 0$. In fact, as L_C is reduced below L_S, larger fraction of tubes bridge source and drain directly, increasing the ratio $I_{\parallel}/I_{\rm random}$ (Fig. 2(a), $\theta_{\rm avg} = 0$). Here, $I_{\parallel}$ and $I_{\rm random}$ are drain current of the networks in Fig. 1(a) and (e), respectively. In the absence of intertube coupling, the optimum angle, therefore, would always be zero (i.e., fully aligned, $\theta_{\rm opt} = 0$). In general, with finite tube–tube coupling, tubes which are not bridging source and drain directly also contribute to I_D by providing percolating paths and this shifts the optimal alignment away from zero ($\theta_{\rm avg,opt} > 0$). However, since higher fraction of tubes directly bridge the source and drain at smaller L_C, contribution from multistick percolation reduces quickly and, $\theta_{\rm avg,opt} \rightarrow 0$ (or more alignment) is needed to optimize smaller L_C transistors [Fig. 2(a), see arrows]. Our simulations show that the gain in the current for optimally aligned network over random network increases with decreasing L_C. In addition, in the limit $L_C \ll L_S$, completely aligned network is most optimal and the current is almost $\sim$100% higher than the random network.

Fig. 2(b) shows the comparison between predicted theoretical estimates and corresponding experimental results to conclusively establish the improvement in the performance of an aligned network [Fig. 1(a)] over a random network [Fig. 1(e)] as a function of L_C/L_S. Broadly speaking, for $L_C \ll L_S$, we find that $\theta_{\rm avg,opt} = 0$, i.e., perfectly aligned network is optimal and the current gain increases from $G \sim 20\%$ for $L_C > L_S$ to $G \sim 100\%$ for $L_C \ll L_S$, a *fivefold* increase for all densities above the percolation threshold $D > D_{\rm perc}$. Here, G is the percentage change of $I_{\parallel}$ over corresponding $I_{\rm random}$. Simply put, better alignment correlates with better I_D performance in short-L_C transistors. This gain in I_D comes at the expense of reduced $I_{D,\rm on}/I_{D,\rm off}$ ratio that saturates to $\sim$3–5 as metallic CNT shorts the S/D of short-channel transistor. Chemical [21] or electrical [4], [6], [22] filtering of multiwalled CNT may be used to restore $I_{D,\rm on}/I_{D,\rm off}$ ratio.

In summary, we have used novel fabrication, characterization, and modeling to consistently interpret experiments involving NB-TFTs with partially/fully aligned nanotube networks. We demonstrate the theoretical limits of gain in ON-state current (20%–40% for long-L_C versus 100% in short-L_C devices) and possibility of fully predictive simulation-guided optimization of transistor performance as a function of density, alignment, and length of CNTs.

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