Large scale, highly ordered assembly of nanowire parallel arrays by differential roll printing

Roie Yerushalmi, Zachery A. Jacobson, Johnny C. Ho, Zhiyong Fan, and Ali Javey[a)]
Department of Electrical Engineering and Computer Sciences, University of California at Berkeley and Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

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A differential roll printing strategy is developed to enable large-scale and uniform assembly of highly aligned and ordered nanowire arrays on various rigid and flexible substrate materials. The dynamics of the process are explored by tuning the linear sliding motion of the roller with respect to the rolling motion, clearly demonstrating the importance of the differential rolling process in the controlled assembly of nanowires. The potency and versatility of the method is further demonstrated by fabrication of nanowire transistor arrays on flexible substrates. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2813618]

Synthetic nanomaterials,[1–3] such as nanowires (NWs), offer unique properties arising from their low dimensional, crystalline structures, and have been proposed as the building blocks for a wide range of technological applications.[1–11] Scalable and controlled assembly of NWs, however, still presents a major challenge toward their potential integration for electronic circuitry.[12–15] Recently, we demonstrated the highly ordered assembly of NW materials using contact printing in which NWs are controllably transferred from a planar growth substrate to a receiver substrate.[16] To extend this process as a generic approach for scalable and large area printing, we demonstrate here assembly of highly ordered, dense, aligned, and regular arrays of NWs with high uniformity and reproducibility using differential roll printing (DRP). We demonstrate that our approach enables large-scale integration of NW arrays on a wide range of substrates including Si, glass, plastic, and paper for electronic applications.

Our approach (Fig. 1) is based on (1) the growth of crystalline NWs on a cylindrical substrate (i.e., roller) using the vapor-liquid-solid (VLS) process, and (2) the directional and aligned transfer of the as-grown NWs from the donor roller to a receiver substrate via DRP. Glass or quartz tubes (outer radius, $r_R \sim 0.25$ in.) were used as the cylindrical growth substrates. The tubes were immersed in $\sim 0.1\%$ w/v of poly-*L*-lysine in water (Ted Pella, Inc.) for 5 min. Subsequently, the tubes were immersed for 2 min in a gold colloid solution. NWs were then grown using the previously reported VLS process.[1] This approach enables uniform growth of dense NW "lawn" (randomly aligned) on the outer surface of the tubes [inset of Fig. 1(b)]. A growth tube was then connected to a pair of wheels and used as the roller of a DRP setup (Fig. 1). A standard I-shape aluminum beam was used as the rail and stage support for the rolling process. The receiver substrate was patterned using photolithography to define the assembly regions and mounted on the stage. The roller was brought into contact with the stationary receiver substrate and rolled with a constant velocity of ~ 5 mm/min, during which NWs are detached and transferred to the receiver substrate. The printing outcome was independent of the rolling velocity for <20 mm/min. At higher velocities, nonuniform printing was observed. The roller-receiver substrate pressure was adjusted to ~ 200 g/cm^2 by a spring underneath the stage. At lower pressures, aligned transfer of NWs was not observed, while higher pressures induced mechanical damage to NWs, resulting in an assembly of short NWs (<1 μm long). Octane was used as a lubricant to minimize the NW-NW mechanical friction during the printing process.[16] Following the NW DRP process, the patterned resist is removed by a standard lift-off process, leaving behind

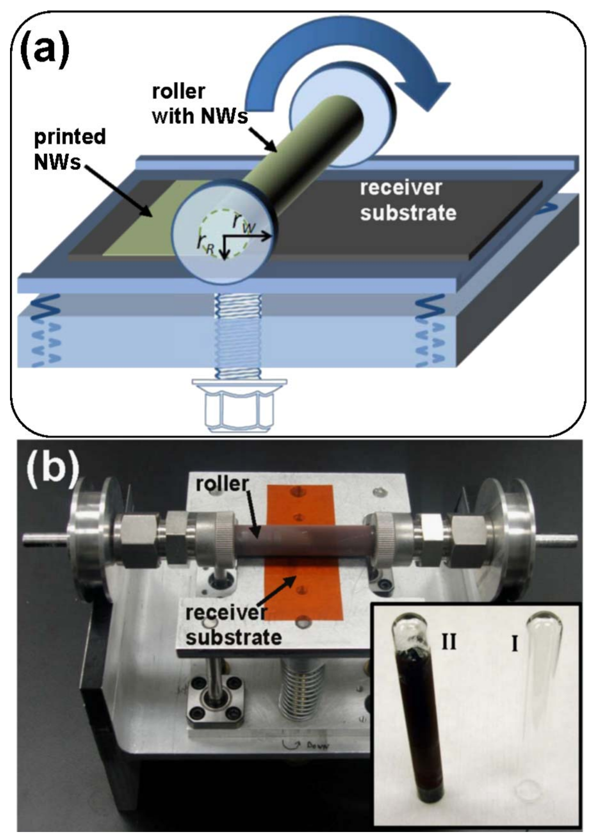

FIG. 1. (Color online) Differential roll printing. (a) Schematic of the DRP setup. (b) Optical photograph of the assembled apparatus used in this work (top view). The inset shows the blank and NW coated glass tubes used as the rollers (I and II, respectively).

a)Electronic mail: ajavey@eecs.berkeley.edu

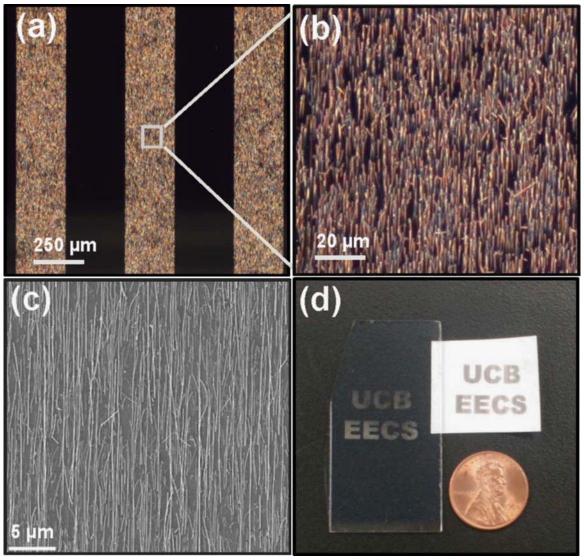

FIG. 2. (Color online) Parallel arrayed NW printing. (a)–(b) Optical dark field and (c) SEM images of GeNWs printed on a Si/SiO_2 substrate. (d) Large area and aligned arrays of GeNWs printed on glass and photography paper. The substrates were patterned using photolithography and functionalized with poly-L-lysine (see Ref. 16) before printing to enable patterned assembly of NWs (shown as the gray regions).

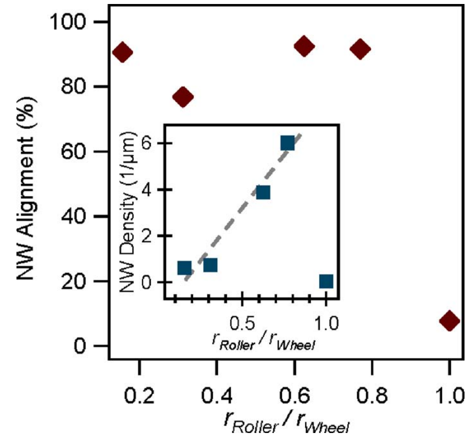

FIG. 3. (Color online) NW printing modulation by wheel size ratio. The percentage of the aligned NWs for various roller to wheel size ratios. The inset shows the NW density as a function of the wheel size ratio.

assembled NWs at the predefined locations. Optical and scanning electron microscopy images of the roller printed GeNWs ($d \sim 30$ nm) on a Si/SiO_2 substrate are shown in Figs. 2(a)–2(c), clearly demonstrating the patterned assembly of well aligned and dense (~ 6 NW/μm) NW parallel arrays. Importantly, the DRP process is compatible with a wide range of rigid and flexible substrates, including Si, glass, plastic, and paper [Fig. 2(d)].

An important aspect of the printing method described here is the mismatch of the roller and wheel radii (r_R and r_W, respectively) that results in a relative linear sliding motion of the roller relative to the stationary receiver substrate in addition to the rolling motion. This is different from traditional roll printing methods where such a mismatch would be undesirable since it would cause a distortion of the printed features. The relative sliding motion for $r_W \neq r_R$ generates the required directing field and shear force for the transfer of aligned NWs to the receiver substrate, without which negligible density with random alignment is observed. To shed light on the NW transfer mechanism, we examined the role of the linear sliding motion on the printing outcome by using different wheel radii. Figure 3 depict the printed NW density and alignment as a function of r_R/r_W. While for $r_W = r_R$, we observe only a small number of NWs (~ 0.04 NW/μm) transferred to the receiver substrate with near random alignment, efficient transfer of NWs with a high degree of alignment ($\sim 90\%$) is observed for all wheel ratios where $r_W \neq r_R$ (Fig. 3). A NW is considered misaligned if it forms an angle $> 5°$ with respect to the direction of printing. The alignment results indicate that it is sufficient to induce a relatively small degree of differential rolling motion in order to attain aligned transfer of NWs, which is expected from the microscopic length of the NW (~ 30 μm) as compared to the macroscopic diameter of the roller. This is consistent with the proposed transfer mechanism of our recently reported contact printing methodology,[16] in which NWs of a planar growth substrate are dragged on a receiver substrate, therefore, effectively aligning them in the direction of sliding. NWs are eventually detached from the growth substrate as they are anchored to the receiver substrate by van der Waals forces.[16] The process is self-limiting with the transfer of NWs limited to a submonolayer without aggregation due to the weak NW-NW interactions [Figs. 2(b) and 2(c)]. For $r_W \neq r_R$, the density of the printed NWs shows a near linear dependence on r_R/r_W (Fig. 3) which is expected since the total number of NWs available for transfer is given as $(2\pi r_R)WN$, where W is the contact area width and N is the NW density of the roller substrate. The printed area covered per revolution is $(2\pi r_W)W$. Therefore, the maximum printed density is given as $N(r_R/r_W)$. From the slope of the printed NW density versus r_R/r_W, we obtain $N \sim 9$ NW/μm, in agreement with the SEM observations of the rollers.

As compared to our previously reported contact printing methodology,[16] DRP minimizes the contact area between the donor and receiver substrates because the cylindrical donor substrate rolls over the receiver substrate with only a small tangent contact area consisting of fresh NWs at any given time. This is highly beneficial for printing large areas that would otherwise require large planar growth substrates and long contact-sliding distances.

To characterize the roll printed NWs, field-effect transistors (FETs) were fabricated on a flexible Kapton substrate using Ge/Si core/shell[9] (15/5 nm) parallel array NWs as the channel material. Figure 4 shows the transfer characteristic of a representative FET fabricated on $a \sim 250$ μm wide NW film with source/drain (S/D) spacing of ~ 3 μm. The transistor delivers ~ 700 μA of on current, corresponding to a semiconductor channel consisting of ~ 700 parallel array NWs, as a single NW delivers ~ 1 μA in the presented device configuration.[16] The estimated NW density from the electrical measurements is ~ 3 NW/μm which is consistent with the SEM observations, indicating that the transferred NWs preserve their intrinsic electrical properties during the DRP process. The transistor performance can be readily enhanced through device geometry optimization[9] and by the use of high mobility NW materials.

In summary, a differential roll printing approach was developed to directly transfer semiconductor NWs from cylindrical donor substrates to patterned receiver substrates. The transferred NWs are highly ordered and aligned. In the future, the process may be extended to enable roll-to-roll printing of NWs for cost-effective, large area electronics.

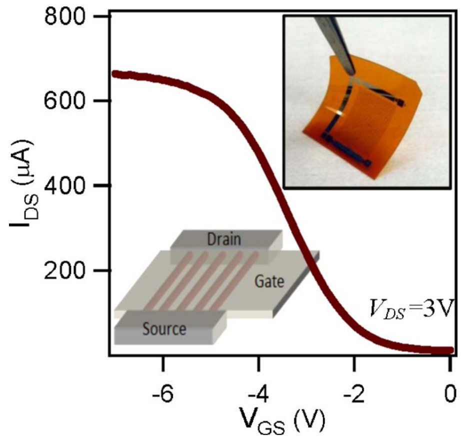

FIG. 4. (Color online) Parallel arrayed NW FETs. *I-V* characteristic of a representative parallel arrayed NW FET with Nickel S/D and G metal contacts and a top-gated configuration ($t_{ox} \sim 15$ nm HfO_2 by atomic layer deposition) on a plastic substrate. The bottom inset shows the schematic of the NW device structure and the top inset is a photograph of the substrate after NW device fabrication.

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