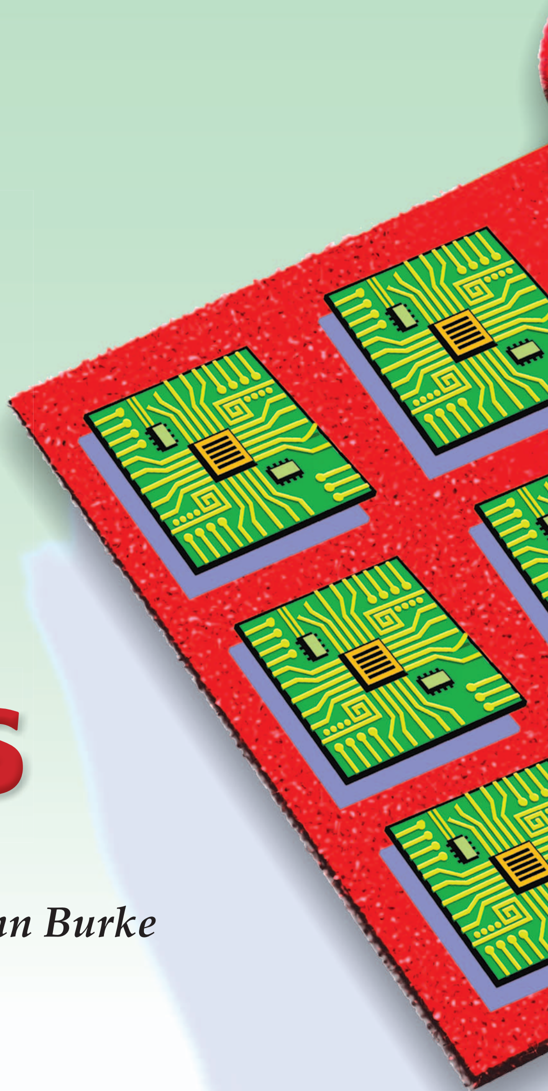
Nanoscale Devices for Large-Scale Applications



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Carbon nanotubes (CNT) and semiconducting nanowires have demonstrated outstanding potential for a wide range of applications, including field effect transistors (FETs) and high-frequency electronics, chemical sensors, nanoelectromechanical systems (NEMS), thin film transistors, and display electronics. Their small size, high mobility (near-ballistic electron transport), high intrinsic cut-off frequency, mechanical and thermal stability, high-current-carrying density, and the capability of having both semiconducting and metallic tubes make carbon-based devices promising candidates to replace silicon-based devices in some applications [1]. One dimensional nanostructures, such as semiconducting nanowires and CNTs, have been heavily investigated as digital devices, but, so far, the study of such structures for radio-frequency (RF) applications is in its infancy. Although the field is still under development, we are beginning to understand some of the possible advantages of such systems over traditional, two-dimensional based technologies. Since 2004, it has been speculated that CNT devices have, potentially, terahertz cutoff frequencies [2]. However, only recently have the device cutoff frequencies crossed into the gigahertz range, as reviewed in [1].

FETs based on dense, parallel arrays of nanotubes are currently being investigated in both academic and industrial laboratories [1], [3]–[7]. To achieve the best possible performance, it is required that a uniform and aligned dense array of all semiconducting nanotubes are on the substrate. Otherwise, parasitic capacitance degrades the cut-off frequency [8]. The presence of

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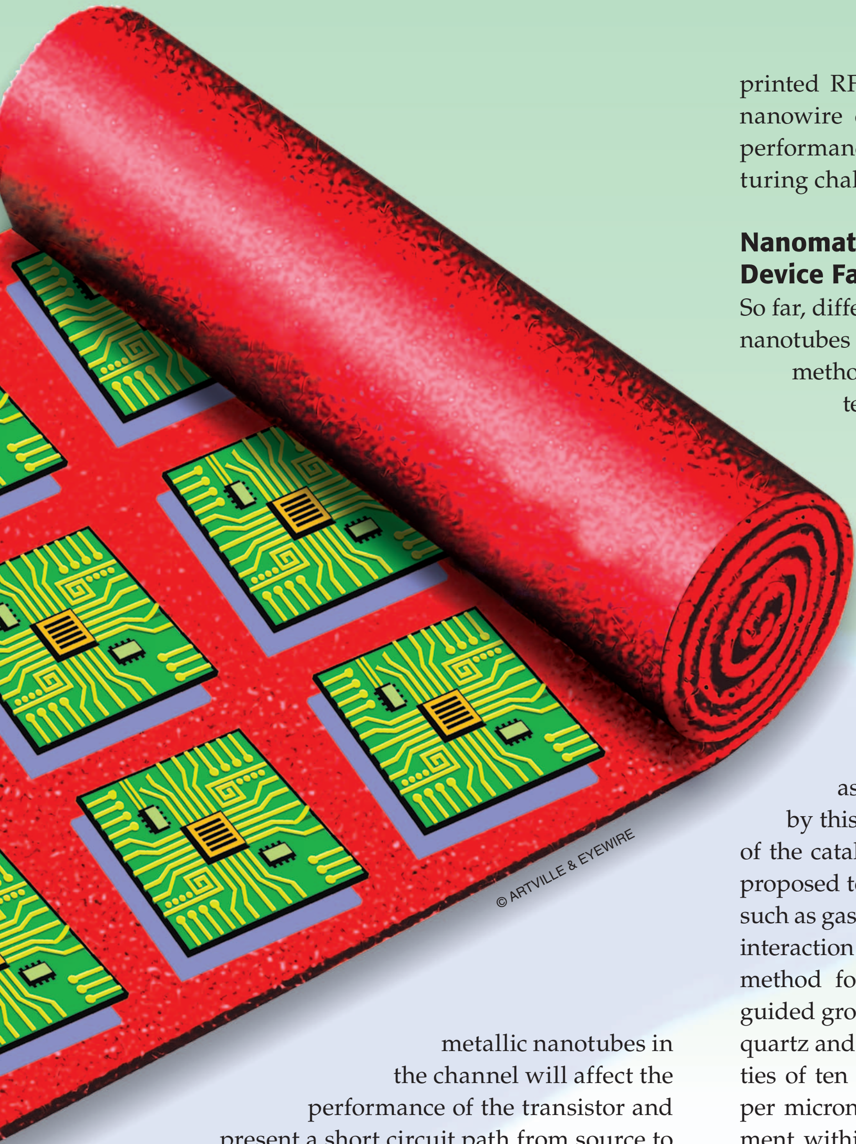

metallic nanotubes in the channel will affect the performance of the transistor and present a short circuit path from source to drain electrode and, as a result, increase the off current and power consumption. Therefore, finding a technique that leads to the mass manufacturability of dense, aligned, and purified semiconducting nanotube networks appears to be an essential step towards RF nanoelectronics. Despite the fact that, so far, it is uncertain whether perfectly dense and aligned arrays containing only semiconducting nanotubes needed for highest performance may be economically manufactured, it is critical to consider state-of-the-art results that have used less-than-perfect nanotube networks. Despite being far from the theoretical limits of individual nanotubes, such results show attractive achievements and great improvements in device characteristics, as well as new horizons for applications in nanoelectronics, including large-area printed electronics [that is, RF identification (RFID) tags] and flexible substrate systems.

In this article, we review the progress so far toward carbon-nanotube-based electronics and consider the implications for application of nanotubes in analogue RF devices and RF system applications as well as recent prospects in printed electronics, especially printed RFID tags. Although our focus is on CNTs, nanowire devices are also starting to show similar performance in the RF domain, with similar manufacturing challenges.

Nanomaterials in Device Fabrication

So far, different techniques have been used to integrate nanotubes in device fabrication. To categorize these methods, one can separate them into two major techniques: 1) in-situ growth of nanotubes and 2) depositing nanotubes.

Grow in Place

The most commonly used technique to grow CNTs is chemical vapor deposition (CVD). The basic idea is to have metal catalysts [iron (Fe), nickel (Ni), cobalt (Co), molybdenum (Mo)] patterned on the substrate and put the sample in a furnace with a flow of carbonaceous gas at temperatures as high as 900 °C. The diameter of nanotubes grown by this method strongly depends on the grain size of the catalyst material. Several techniques have been proposed to obtain aligned and dense nanotube arrays such as gas flow [2], [9]–[11], electric field [12], [13], and interaction with the substrate [1]. The most consistent method for gaining an aligned network is surface-guided growth on single-crystal (SC) substrate, such as quartz and sapphire [1], [5], [14]–[18]. Nanotube densities of ten tubes per micron (peak value of ~50 tubes per micron), lengths of more than 100 μm and alignment within <0.01° were obtained using this method [Figure 1(a) and (c)] [1]. Moreover, nanotubes grown by this method can be transferred to any arbitrary substrate, as described in [19], which makes this technique suitable for flexible substrates too. Statistics show that nanotubes grown using the CVD method typically contain two-thirds semiconducting and one-third metallic tubes. This ratio of semiconducting single-walled nanotubes (s-SWNTs) to metallic SWNTs (m-SWNTs) affects the device performance, especially the on/off ratio. Off currents are related to only the metallic tubes and, having one-third metallic tubes, degrades the on/off ratio below 100, which is very low when compared to conventional electronic devices. The low on/off ratio corresponds to high standby power dissipation. Meanwhile, experimental studies show that using individual semiconducting nanotubes for the channel of FET results in an on/off ratio of more than 10^6, but this ratio is four to five orders of magnitude lower for the s-SWNT and m-SWNT mixtures.

Different techniques are used to decrease the number of metallic tubes, either during the growth process or after, as reviewed in [1]. Most of these methods require individual access to the nanotubes (such as

Carbon Nanotube

Carbon nanotube is an allotrope of carbon which is theoretically described as a folded single sheet of carbon, called graphene. Depending on where the cut through the sheet is formed, the final nanotube will have different band-gap and therefore various electrical properties. Based on the cut, there are generally 3 different types of nanotube; Armchair, Zigzag and Chiral. Also the (n,m) index defining the orientation of the cut is called the chirality of the tubes. Based on the number of coaxial tubes forming the nanotubes, they are classified as Single-Walled (SWNT), Double-Walled (DWNT) or Multi-Walled nanotubes (MWNT) each has distinguished properties for electrical and chemical applications.

Because of the sp^2 bonding (stronger than sp^3 in Diamond) and also the unique molecular arrangement of nanotubes, they play an interesting role in chemical sensors as well as nanomechanical systems. In addition, the band-gap properties and 1-dimentional nature of nanotubes puts them among the promising candidates in future nanoelectronics. The near-ballistic electron transport in nanotubes makes it possible to obtain very high mobility circuits out of an individual nanotube. Moreover, the low quantum capacitance gives it an intrinsic frequency operation in the range of THz with the ability of high current carrying. These unique properties make carbon nanotube an interesting material to explore for future applications in electrical, chemical and mechanical systems.

electrical breakdown [20]), which is not feasible in high transistor densities. Alternately, post processing may be applied, which may also affect the semiconducting nanotubes and, therefore, the performance of the whole device [21], [22].

Although CVD growth of nanotubes may give the option of aligned arrays along the substrate crystalline structure, growing nanotubes in place may cause contamination such as catalyst and amorphous carbon contamination that can drastically affect the device. Also, the temperature for CVD growth is beyond the tolerance of most of flexible substrates. Growing aligned, purely semiconducting tubes is something that is not feasible to date, which makes this technique unsuitable for high-frequency, high-mobility, low on/off ratio CNT FETs.

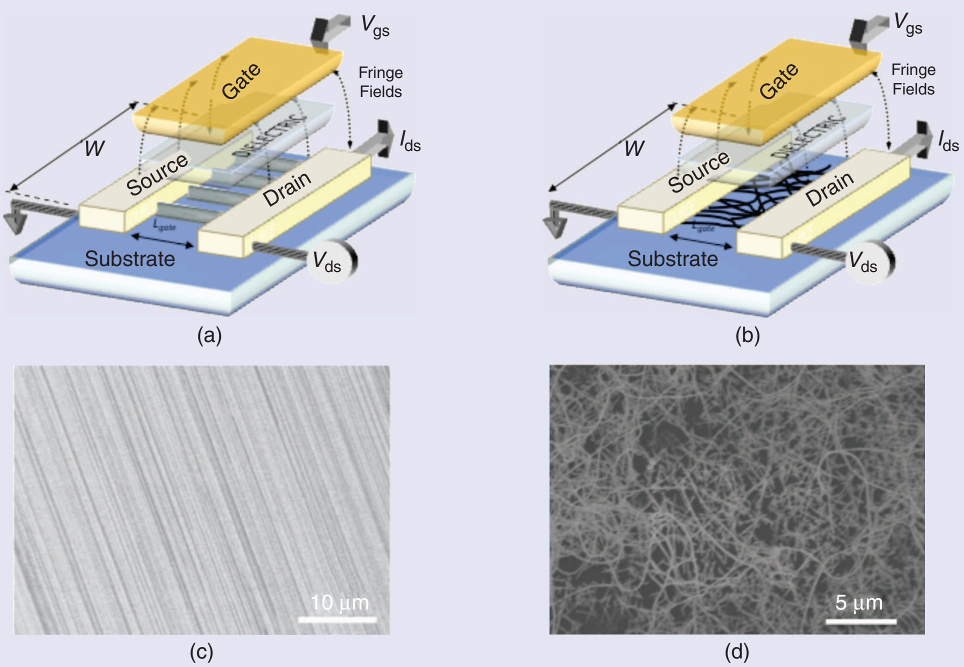

Figure 1. *(a) Schematic of top-gated carbon nanotube field-effect transistor using aligned chemical-vapor-deposition-grown nanotubes on a single crystal-quartz substrate [1]. (b) Nanotube field-effect transistor using solution-based deposition of carbon nanotubes. (c) Scanning electron microscopy image of chemical-vapor-deposition-grown nanotubes on single crystal-quartz [5]. (d) Scanning electron microscopy image of random network of nanotubes deposited from a semiconducting-enriched solution [31].*

Solution-Based Deposition

Depositing nanotubes from a solution has also been investigated recently to find the best way of producing an aligned and dense network of semiconducting nanotubes [23]–[25]. Several methods such as selective chemistry [26], [27] and dielectrophoresis (DEP) [28] were used to deposit nanotubes from a solution. The first one is based on chemical molecules attached to the nanotube in the solution and then separated by the density gradient of the nanotubes in the solution. The latter method uses dielectrophoretic force to separate the metallic from semiconducting nanotubes. Main challenges in the naotube deposition method are

- finding an economical process
- sorting large diameters
- obtaining longer nanotubes in the solution (especially for long channel lengths)
- studying the influence of the residual surfactants.

A deposition technique to get aligned nanotubes in a large area should be considered as well. Along with using nanotube solutions enriched in semiconducting nanotubes, modification of the substrate surface prior to the deposition is another method that is currently being investigated for having selective placement of nanotubes on wafers [23], [24], [29]. Amine terminated silanes, such as 3-aminoprolyltri (ethoxysilane), provide far better and selective deposition of semiconducting SWNTs when attached to the substrate surface [23]. Such modifications enhance the adsorption of nanotubes from most of the aqueous dispersions of nanotubes.

Figure 1 shows schematics and images of two different approaches for nanotube device fabrication. The solution-based deposition technique involves high-speed spinning of the wafer and dripping a solution of nanotubes onto it. The result shows radially aligned nanotubes. Recent investigation showed that using the spin-coating method for nanotubes on a modified surface, as stated above, can enhance the on/off ratio to more than 10^5, while putting a droplet on the wafer and then spinning the wafer may degrade the on/off ratio by up to three orders of magnitude [30]. Densities of more than ten tubes per square micron were achieved using this method [30]. To date, all efforts were successful in improving one of the features, either the on/off ratio or the mobility, separately. However, to be able to integrate nanotube devices in electronic systems, it's desirable to have a high-mobility transistor and high on/off ratio at the same time. Only recently was it shown that combining the two fabrication methods described above could help in achieving both goals. Using all semiconducting enriched nanotubes that are chemically separated and also surface modification to absorb semiconducting nanotubes on the surface can lead us to gain mobilities as high as 40 cm^2V^{-1}-s^{-1} while keeping the on/off ratio higher than 10^4 [31]. This study shows a great improvement in RF device fabrication and shows potential applications in flexible electronics used for RFID tags, which will be discussed later. Although the solution-based deposition of nanotubes is conceivably scalable, the formation of aligned and dense nanotube networks is an interesting future step toward improving the characteristics of the devices.

Finding a technique that leads to the mass manufacturability of dense, aligned, and purified semiconducting nanotube networks appears to be an essential step towards RF nanoelectronics.

Regardless of which method described above is chosen, following the nanotube deposition, fabrication steps are almost the same as conventional techniques used for processing transistors. For large-size patterning, conventional photolithography is used to define the source, drain, and gate electrodes, while for submicron dimensions, the advanced electron-beam lithography is required. After defining the electrodes and channels, e-beam evaporation is mostly used to lay down the metal electrodes followed by a lift-off process. Since nanotubes intrinsically show a p-type (type of semiconductor that has free holes as the majority carriers) behavior due to oxygen absorption on their surface, the best metal known to make an ohmic contact (rather than a Schottky contact) would therefore be Palladium. We use Palladium for the first metal layer, which is in direct contact with the nanotubes. Then gold is used to cover that to make a very-low-resistance electrode on the very top layer.

RF Nanodevices

Using a typical linear circuit model for an RF transistor that consists of a current source (voltage dependent), resistances, and capacitances, the extrinsic cut-off frequency (due to parasitic elements) can be calculated as

$$f_T = \frac{g_m}{2\pi} \cdot \frac{1}{(C_{gs}+C_{p,gs}+C_{p,gd})[(R_{p,s}+R_{p,d})g_d+1]+g_m C_{p,gd}(R_{p,s}+R_{p,d})}, \tag{1}$$

where g_m is the transconductance, g_d is the drain conductance, $C_{p,gs}$ and $C_{p,gd}$ are gate-source and gate-drain parasitic capacitances, and $R_{p,s}$ and $R_{p,d}$ are parasitic series resistances for the source and drain [1]. Ignoring these parasitic capacitances and resistances can give us the intrinsic cut-off frequency

Carbon nanotube transistors are capable of entering the terahertz frequency regime while maintaining high mobility and a high on/off ratio.

$$f_{T-\text{Intrinsic}} = \frac{g_m}{2\pi C_{gs}}. \quad (2)$$

The intrinsic cut-off frequency can also be considered as the highest cut-off frequency of the device; in its best performance, the intrinsic cut-off frequency is typically in the THz regime [8]. Improving the metals and contacts can lower the parasitic resistances. Therefore, the most important parasitic element tends to be the capacitances.

The electronic properties of SWNTs depend on their chirality and diameter. It's been shown that the bandgap of nanotubes is inversely related to their diameter [32]. It's been shown in [1] that, in CNT FETs, C_{gs} is proportional to L_{gate}, therefore, we conclude that the cut-off frequency can be calculated for different gate length regimes as

1) large-channel lengths

$$f_t = \frac{\mu(V_g - V_t)}{2\pi L_g^2} \quad (3a)$$

2) small-channel lengths

$$f_t = \frac{v_{sat}}{2\pi L_g}. \quad (3b)$$

The saturation velocity for CNTs is $v_{sat} = 2 \times 10^7$ cm s^{-1} [33], [34]. Taking this into account, the cut-off frequency for short-gate-length devices will be around 30 GHz/L_{gate} (μm). This is comparable to state-of-the-art III-V devices with the advantage of the nanotube's small size, which allows it to go into deep submicron dimensions. Moreover, with long channel devices, as in printed electronics, mobility plays an important role to obtain higher cut-off frequencies; therefore, the focus of recent studies has been to improve the mobility.

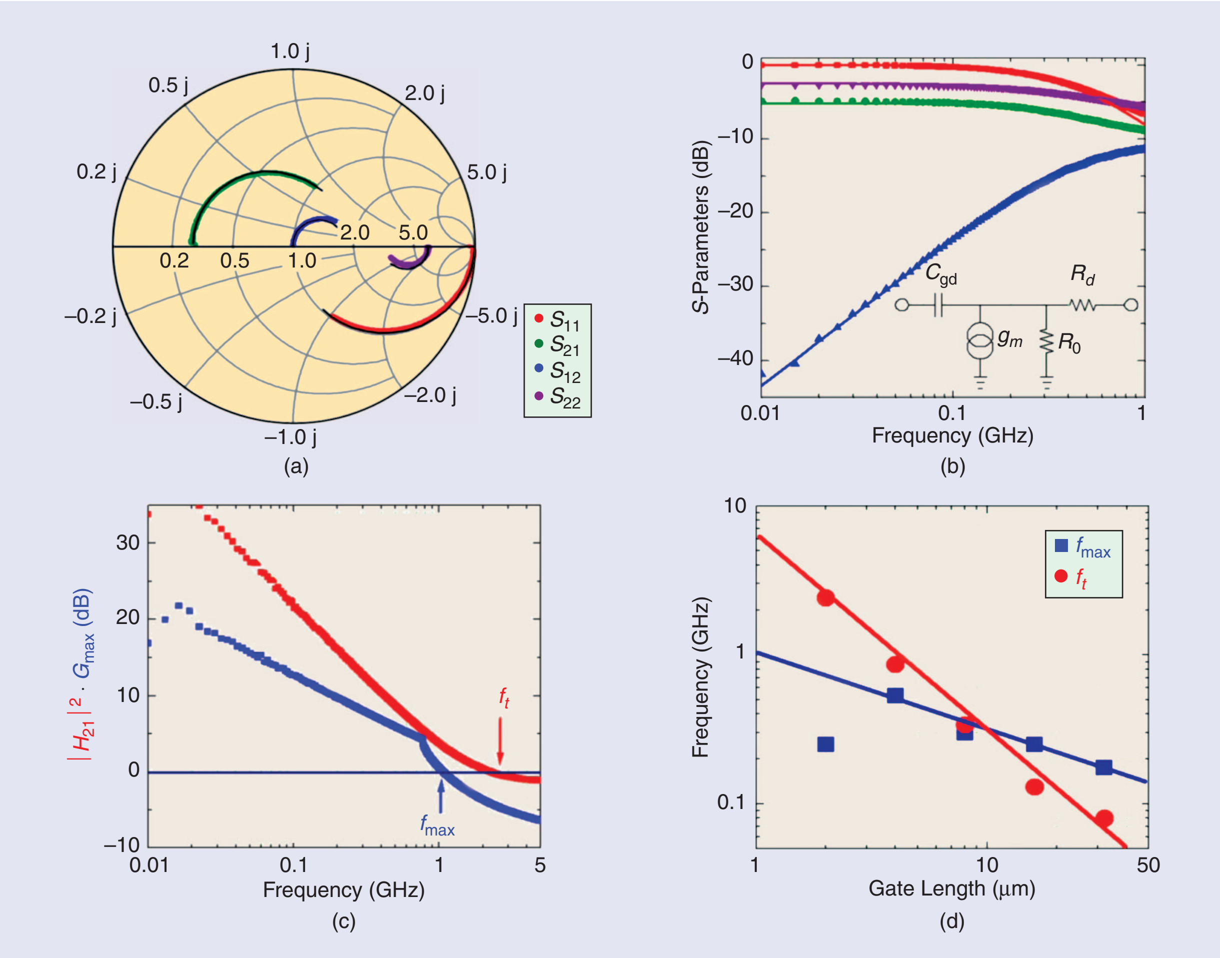

Figure 2. *(a) Smith chart and (b) amplitude curve of measured two-port S-parameters of the device fabricated in [35] ($L_{channel} = 8$ μm and $W = 300$ μm). (c) Current gain and maximum power gain plots showing the f_t and f_{max} ($L_{channel} = 4$ μm and $W = 100$ μm). (d) f_t and f_{max} versus channel length with $W = 300$ μm.*

Cut-Off Frequency

Cut-off frequency is an important figure of merit for any transistor fabricated for electronic circuits. It is basically the frequency at which the current gain becomes 1 or 0 dB and defines the boundary of frequency response for the system. Although in reality the transistor wont keep its ideal characteristics at this frequency, but it is still the highest theoretical frequency beyond which the transistor/system won't give a desirable response.

Mobility

Mobility is a term to characterize the movement of charge carriers (electron or hole) in any material when an electric field is applied. Mobility is based on the scattering properties of the material which itself depends on the physical and molecular/atomic arrangement of that medium.

On/Off Ratio

The ratio of the On-state current to the Off-state current of a transistor is called on/off ratio. In electrical circuits especially in digital systems it is important to have as low off current as possible since the off current corresponds to the static power consumption of the circuit. Therefore, the higher the on/off ratio (that is, the lower the off current) the better the system will work and the lower the power consumption. On/Off ratio is also considered as one of the figures of merit for the transistor.

Working toward a dense and aligned array of nanotubes can help us increase the cut-off frequency closer to its intrinsic value. Increasing the number (density) of nanotubes will increase the transconductance (g_m) without a significant change in the capacitance, thus improving the cut-off frequency.

Figure 2 shows the Smith chart and scattering parameters extracted from FETs built on an SC-quartz substrate with aligned CVD-grown CNTs [35]. Using the scattering parameters, the current gain was calculated for different frequencies to obtain the cut-off frequency. As shown in Figure 2(d), both the cut-off frequency and maximum power gain frequency increase by decreasing the gate length, and the trend shows the possibility to enter the terahertz regime by scaling down the gate length into the nanometer range.

Figure 3 also shows recent results for high cut-off frequency FETs made with semiconducting-enriched CNTs and submicron channel length (around 300 nm). Unlike the results from Figure 2, here solution-based deposition was used to deposit 99% semiconducting nanotubes in the channel. A cut-off frequency of 80 GHz (excluding the parasitic capacitances) was achieved using this method [4].

Nanoelectronic Systems

Nanotube Radio

As described previously, CNT transistors are capable of entering the terahertz frequency regime while maintaining high mobility and high on/off ratio. Recent works have also focused on demonstrating electronic systems using nanotube transistors (Figure 4). However, to be able to enter the commercial market, they need further improvement and scalability.

Nanotube radios were the first demonstration of nanotubes as the demodulator in a radio receiver [7], [36]. Because of their nonlinearity, nanotubes can demodulate an AM RF signal. The nonlinear current-voltage characteristic of nanotubes can rectify the applied RF current, which to first order comes out to be

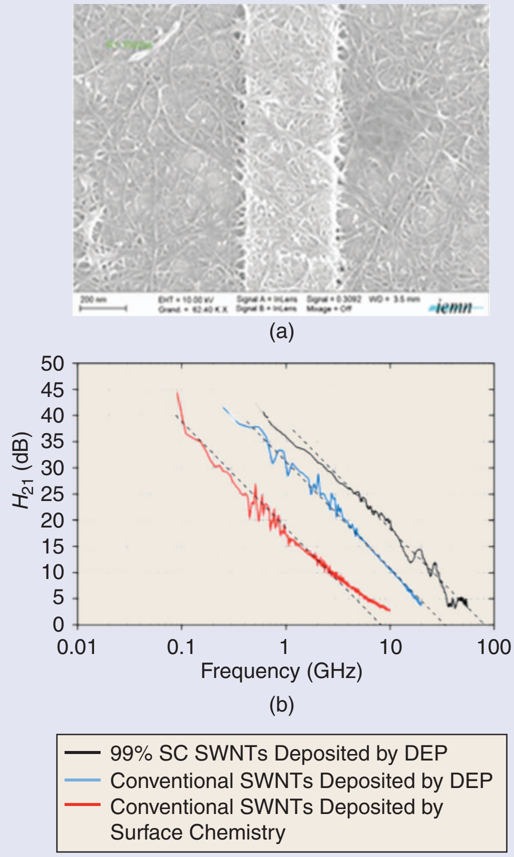

Figure 3. *(a) Scanning electron microscopy image of carbon nanotubes deposited on the gate electrode. (b) Current gain for the three single-walled nanotube field-effect transistor cases with f_t (cut-off frequency) of 8 GHz, 30 GHz, and 80 GHz, respectively [4]. DEP: dielectrophoresis.*

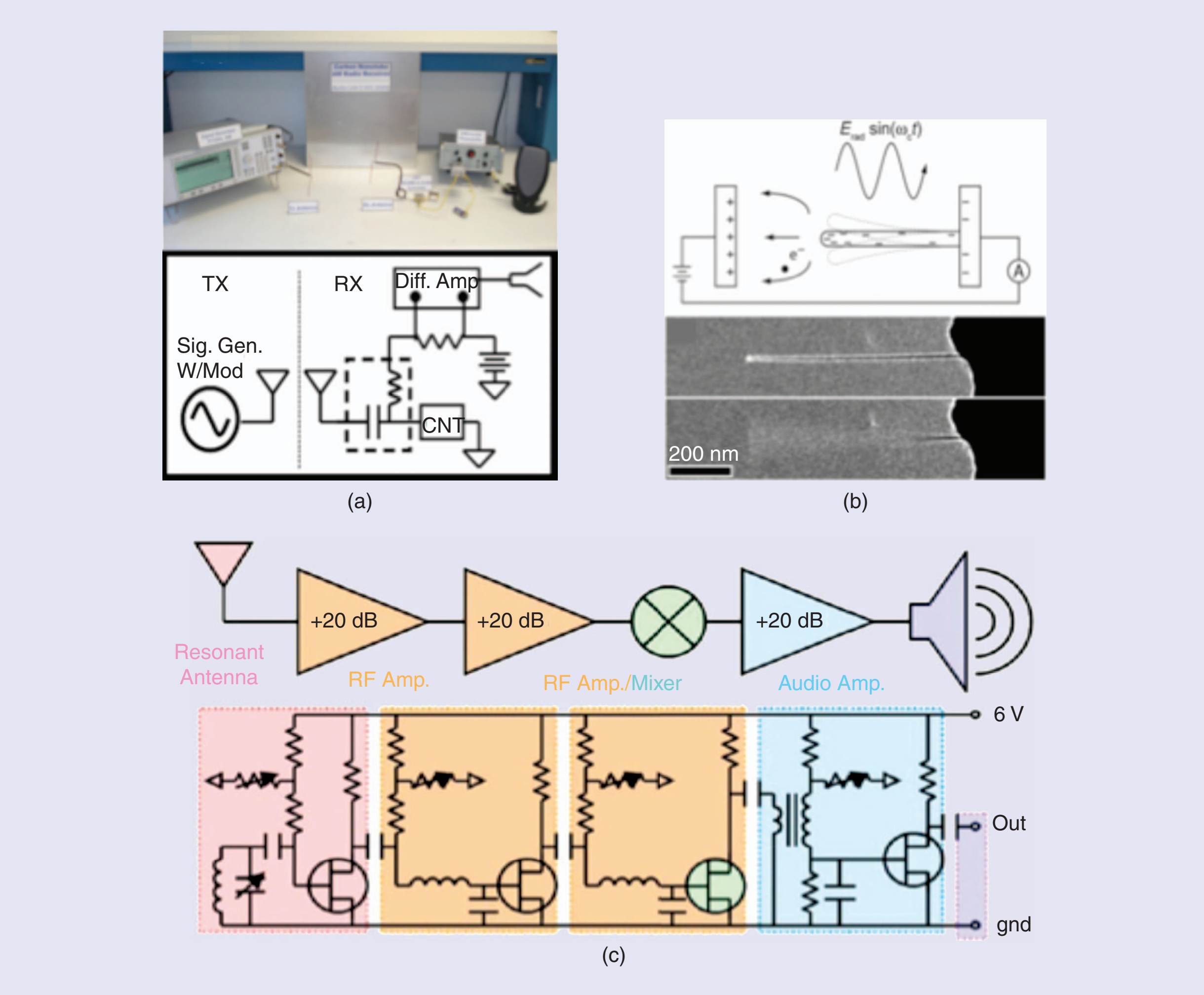

Figure 4. *(a) Schematic of a carbon nanotube radio demonstration [7]. (b) A nanotube in a high vacuum acts as an RF detector and integrated RF filter [36]. (c) Nanotube field-effect transistors act as the RF preamplifier, detector (mixer), and audio frequency amplifier, thus demonstrating a complete AM radio system [35].*

$$I = \frac{1}{4} I_0 V_{\mathrm{RF}}^2 \frac{\partial^2 I}{\partial V^2}, \tag{4}$$

where V_{RF} is the RF voltage signal, and the second derivative represents the nanotube's current-voltage nonlinearity [7].

Although none of the demonstrated nanotube radio systems have been sensitive enough for weak radio signals, they show one promising application of nanotubes in RF systems. Furthermore, other work shows the potential employment of CNTs in narrowband amplifiers operating in the VHF frequency regime with a power gain of 14 dB [35]. Based on the scattering parameter measurements, a cut-off frequency of 2.5 GHz and a maximum power gain frequency ($f_{\max}$) of 1.1 GHz was achieved [Figure 2(c)].

Printed RFID

In the area of flexible substrates, recent achievements show that ink-jet-printed nanotube devices can be used to fabricate RFID tags [37], [38]. An elegant research study used semienriched nanotube ink to print the first all-printed roll-to-roll nanotransistors for RF applications, as shown in Figure 5. However, these devices suffer from low mobility in the range of less than 1 $\mathrm{cm^2\,V^{-1}s^{-1}}$ (while having a high on/off ratio of 1,000) or mobility of maximum ~5.5 $\mathrm{cm^2\,V^{-1}s^{-1}}$ with on/off ratio of 100 [39]. This trade-off can be resolved as described in [29], [31]. Using semiconducting-enriched nanotube solutions and depositing them on a premodified surface can ensure the deposition of more than 90% semiconducting CNTs. Even though the nanotubes will be deposited in random directions, the results demonstrate a drastic improvement in mobility and on/off ratio and provides scalability for ink-jet printing of nanotubes on any arbitrary surface. Recent results show the ability to obtain a high mobility as well as high on/off ratio in the order of 10,000 [31]. Based on the current-voltage and depletion curves,

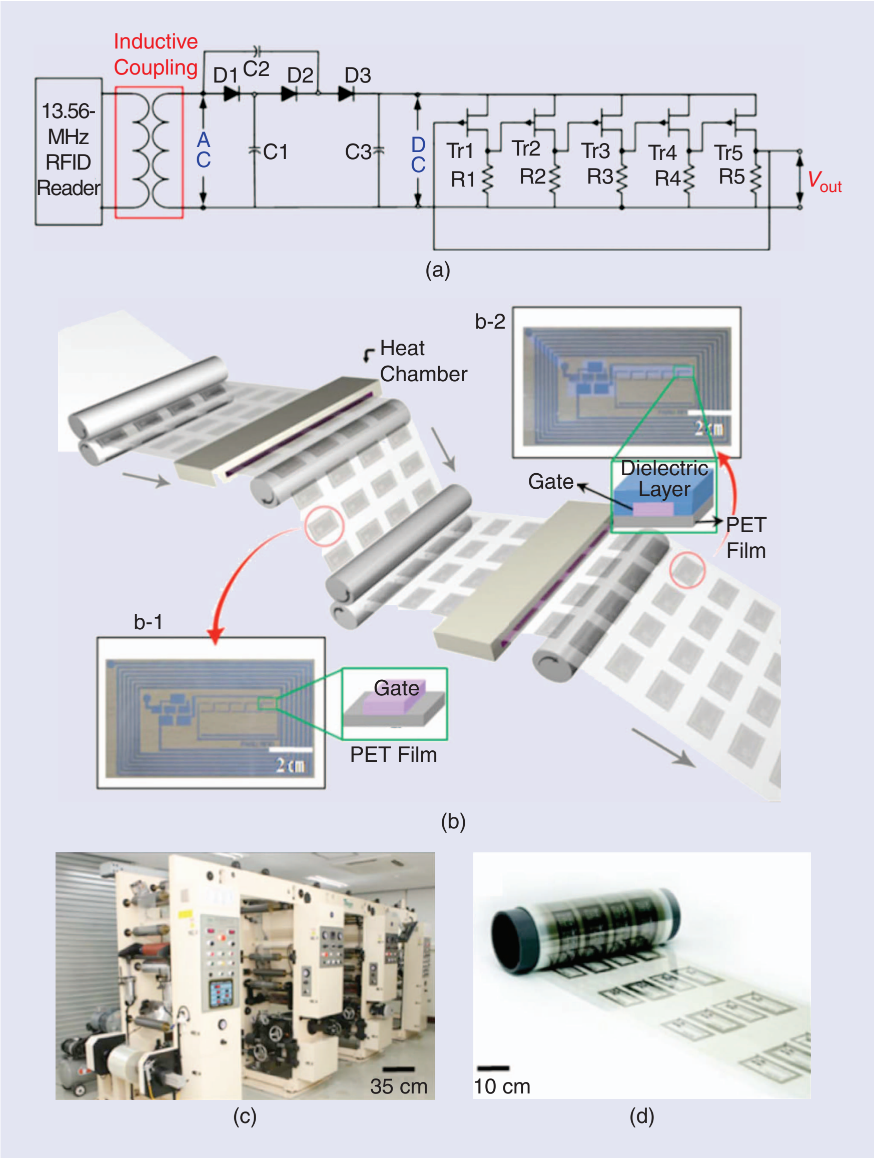

Figure 5. *(a) Schematic circuit diagram for a 13.56-MHz-operated 1-bit RF tag. (b) Schematic illustration of roll-to-roll gravure-printed antennas, electrodes, and wires after passing through the first printing unit using silver-based conducting ink (b-1). Printed dielectric layers on selectively designated spots using high-K dielectric ink after passing through the second printing unit (b-2). (c) Gravure printer used in this work with two printing units. (d) Roll image of the completed roll-to-roll gravure-printed 13.56-MHz antenna, electrodes, wires, and dielectrics used as precursor for printing 13.56-MHz-operated 1-bit RF tags [39].*

one can use the traditional CMOS equations to find the mobility.

$$I_{ds} = \mu C \frac{W}{L}\left[(V_{GS} - V_t).V_{ds} - \frac{1}{2}V_{ds}^2\right]. \qquad (5)$$

Mobilities as high as 40 cm^2V^{-1}-s^{-1} were achieved with an on/off ratio of 10,000 in the same device. Being able to improve both mobility and on/off ratio encourages us to implement this technique in the ink-jet printed electronics, as shown in Figure 5.

Despite the demonstration of nanotube transistors integrated in RF systems, it is still necessary to make all other components of these systems, such as power supplies, antennas, amplifiers, signal processing devices, and mixers, out of nanotube devices to have a fully integrated RF nanosystem. Further investigation is essential to come up with a commercial protocol that is capable of replacing CMOS, III-V, and flexible electronics technology in terms of efficiency, power dissipation, frequency operation, and size of the devices/systems.

Summary

Different CNT synthesis methods have been studied in this article along with an RF circuit model for FETs. An analytical review of individual nanotubes leads to better understanding of various parameters influencing the device's performance such as cut-off frequency, mobility, and on/off ratio. It has been shown that, to achieve high-performance, high-frequency carbon-based transistors, it is essential to have purified all-semiconducting nanotubes densely aligned in the channel. A promising option would be using a nanotube solution deposited on the substrate.

Recent achievements in the area of RF nanotube systems, including RF nanotube radios and amplifiers, also have been discussed. New ink-jet printing techniques open up great possibilities for printed RFID tags in the future. An important milestone in RF device/system fabrication would be to obtain both high mobility and a high on/off ratio. Current research works are focusing to improve all these properties at the same time.

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