Alignment of carbon nanotubes on pre-structured silicon by surface acoustic waves

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Abstract
Carbon nanotubes have been deposited and aligned onto the pre-structured metal contacts of a silicon chip. Crucial for the deposition and alignment process are micro-fluidic flow fields combined with electric dipole fields generated by surface acoustic waves within a gap filled with an aqueous carbon nanotube suspension. This gap is formed when the pre-structured silicon chip is flipped onto the piezoelectric lithium niobate substrate, allowing for the generation of surface acoustic waves. The contacting probability of carbon nanotubes on the prestructured metal contacts has been found to be 37%. In combination with back-gates, these structures define three-terminal devices and the first current–voltage characteristics.

1. Introduction

Since their discovery in 1991 [1], carbon nanotubes (CNTs) have been attracting great scientific interest. Carbon nanotubes are widely regarded to be promising candidates as building blocks for future nanoelectronic devices [2, 3], field emitters [4], sensors [5], nano-electromechanical systems [6] and also quantum devices [7, 8].

The lateral manipulation and control of location and orientation or the pristine growth of carbon nanotubes with specific electronic or mechanical properties is therefore considered to be a key for the breakthrough of carbon nanotube applications. In this respect, several different approaches have been reported so far, such as the lateral manipulation of carbon nanotubes using the tip of an atomic force microscope [9, 10], or the deposition of carbon nanotubes mediated by the application of electric fields [11], magnetic fields [12, 13], or the pre-patterning of substrates with catalysts [14, 15]. Mechanical fields, generated by gaseous nitrogen flowing from a nozzle, have also been used to align carbon nanotubes onto substrate surfaces [16].

Recently, we have demonstrated the deposition and alignment of multi-walled carbon nanotubes (MWNTs) and single-walled (SWNTs) carbon nanotubes onto the surface of piezoelectric substrates like lithium niobate or lithium tantalate using surface acoustic waves [8, 17].

Due to the overarching relevance of silicon in the development of microelectronics in the last five decades, we developed a method to deposit and align SWNTs and MWNTs onto pre-structured silicon.

2. Method

The carbon nanotubes are dissolved in water mediated by the surfactant SDS (sodium dodecylsulphate) at the critical micelle concentration [18, 19]. The approach to deposition from an aqueous carbon nanotube suspension instead of growing carbon nanotubes directly by chemical vapour deposition can be considered to be favourable, as successful experiments to separate semiconducting SWNTs from metallic SWNTs in an aqueous SDS-mediated carbon nanotube suspension have already been reported [20].

The purified SWNT raw material was obtained from Tubes@Rice, Houston, USA, while the MWNT raw material was purchased from ILJIN Nanotech Co. Ltd, Seoul, Korea.

A flip-chip setup, as shown in figure 1, has been used to exploit the favourable properties of surface acoustic waves (SAW) generated on the piezoelectric substrate lithium niobate

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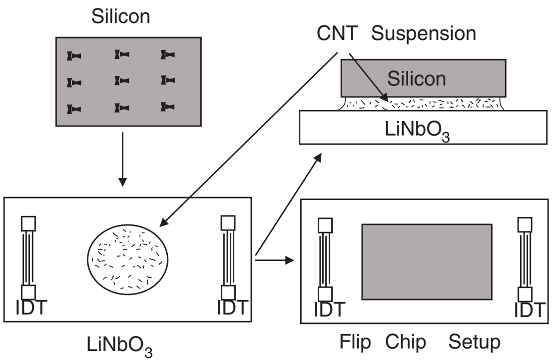

Figure 1. Flip-chip setup for deposition and alignment of carbon nanotubes (CNTs) on a pre-structured silicon chip using surface acoustic waves. Not to scale.

(This figure is in colour only in the electronic version)

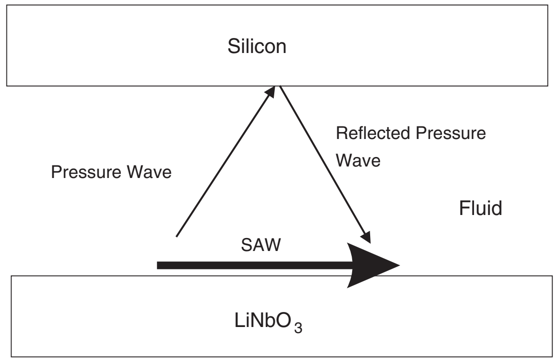

Figure 2. Rayleigh waves couple into a thin fluid film in between the piezoelectric substrate and a cover slide and are back reflected at the cover slide to again reach the piezoelectric substrate.

in combination with a non-piezoelectric silicon chip. The crystal orientation of the lithium niobate substrate, the tool chip, is rot128°-Y-cut-X-propagation. A pair of metallic interdigital transducers—forming a delay line—has been processed by standard photolithography and fabricated by evaporating a 10 nm thick titanium adhesion layer and a 200 nm thick gold layer in combination with a standard lift-off technique (commercially avaible at Advalytix AG, Brunnthal, Germany). The periodicity of the interdigital transducers is 15.2 μm, resulting in a resonant frequency of the SAW on lithium niobate of 262 MHz.

The silicon chip (5×5 mm^2, n$^+$ arsenic doped, resistivity 0.001–0.005 Ω cm, orientation $\langle 100 \rangle$) has been covered with 200 nm of SiO_2 by plasma-enhanced chemical vapour deposition to serve as a gate dielectric for a back-gate contact. Electron-beam lithography (using a Raith 100) has been used to pattern five samples on a chip each with 18 contact pairs (10 nm titanium followed by 50 nm of gold) with a separation of 500 nm. The contact pads had a base width of 200 μm and a 5 μm-wide capped tip, such that two adversely placed contact pads separated by 500 nm are forming the basis of one junction.

In the flip-chip setup (see figure 1), the oxidized silicon chip with pre-structured gold contacts is brought into close proximity to the electric SAW field generated independently on the separate piezoelectric lithium niobate chip.

For the fabrication of carbon nanotube junctions, a chip carrier with a lithium niobate chip has been connected to a signal generator (a Rohde&Schwarz SMP02, 10 MHz–20 GHz) via an RF power amplifier (EIN, Model 603L, 0.8–1000 MHz, +40 dB). The electrical signal detected by the receiving interdigital transducer of the lithium niobate chip has been fed into a spectrum analyser (an Agilent E4402B, 9 kHz–3.0 GHz) and adjusted in power to 7 dBm, being equal to 5 mW. After preliminary power adjustment, the signal generator is switched off and 1 μl of a freshly prepared suspension of carbon nanotubes is pipetted into the middle of the sound beam. A clean pre-structured silicon chip is subsequently flipped onto this droplet. Once the signal generator was in operation, the power level has been adjusted once more to 7 dBm, as measured with the spectrum analyser. Capillary forces spread the droplet of carbon nanotube suspension in between the tool chip and the target chip into a film that gradually diminishes in thickness due to evaporation. The deposition and alignment process was carried out for ten minutes until the aqueous part of the carbon nanotube suspension evaporates completely. To remove residual surfactant, the target chip was rinsed for 30 s in de-ionized water and subsequently dried in a nitrogen gas flow.

3. Results and discussion

Silicon does not allow the excitation of surface acoustic waves directly due to the absence of dielectric dipole moments per silicon unit cell. Thus, the evanescent fields of a SAW in the third dimension above the piezoelectric chip are exerted on the surface of the pre-structured silicon chip. The flip-chip setup, as sketched in figures 1 and 2, facilitate the combination of both micro-fluidic and electric properties of the SAW with a second substrate through close spatial proximity.

SAWs interact with fluids by exchange of energy if the fluid is in direct contact with the sound beam generated on the piezoelectric substrate. Rayleigh waves couple into the fluid and are attenuated exponentially such that acoustic streaming occurs [21]. In the case of a covered piezoelectric substrate shown in figure 2, Rayleigh waves couple to the fluid, where they excite a longitudinal pressure sound wave. This wave is reflected back from the cover lid and propagates along the fluid-filled gap, as in a waveguide.

Thus, micro-fluidic streaming occurs in the narrow gap formed in between the lithium niobate chip and the pre-structured silicon chip.

Additionally, electric dipole fields which accompany a SAW propagating on a piezoelectric substrate play an important role in the polarization and subsequent alignment of carbon nanotubes [17, 22]. In the upper half-space above a piezoelectric substrate, a SAW causes a modulated electric field similar to a periodic surface charge density. Surface acoustic waves on piezoelectric substrates hence originate periodic electric fields of plane-wave nature and exhibit evanescent behaviour in the third dimension.

The combination of both micro-fluidic streaming and the aligning effect of the electric fields facilitate the deposition of carbon nanotubes between the pre-structured gold contacts on the silicon chip.

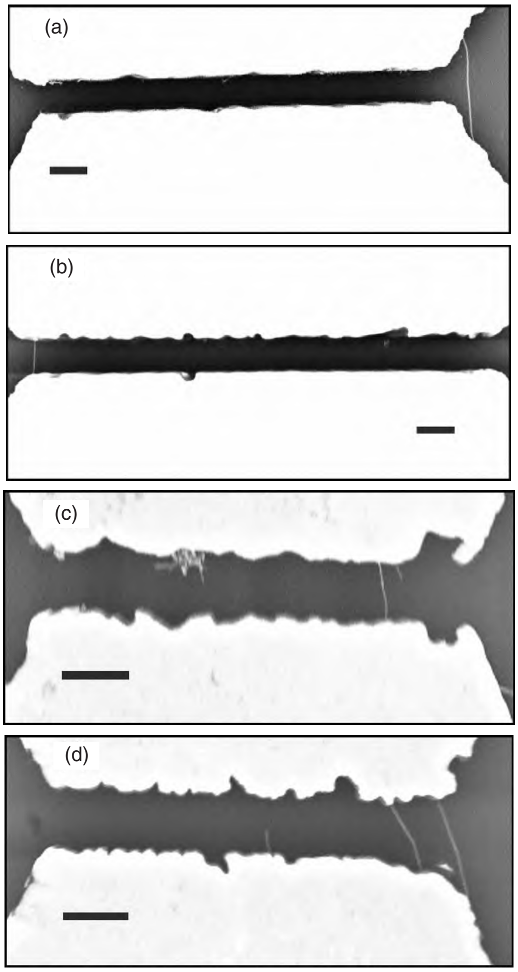

Figure 3. SAW-aligned CNT between a pair of gold contacts pre-structured onto an oxidized silicon chip. In (a) and (b), scanning electron microscopy (SEM) images depicting single-walled carbon nanotubes aligned between pre-structured metal contacts are shown. In (c) and (d), multi-walled carbon nanotubes are shown to contact pre-structured metal pads. For the deposition, the flip-chip setup was used. The bar scale is 500 nm.

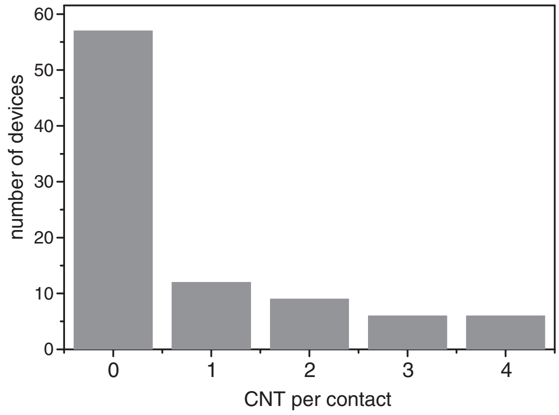

Figure 4. Statistics of the CNT contacting process in the flip-chip geometry. The CNT suspension used for the deposition had a deliberately low CNT concentration. Therefore, most of the inspected contact pairs are free of nanotubes. Almost 37% of the devices, however, result in a CNT junction and most of these are bridged by only a single CNT or two.

Electric field amplitudes between 10^6 and 10^7 V m^{-1} are found to accompany surface acoustic waves on piezoelectric substrates for an amplitude of surface displacement of around 1 nm [23, 24].

For the measurements presented here, relatively high power levels have been used in order to launch SAWs with large mechanical and electrical amplitudes. We used power levels of up to 500 mW (signal generator output), since the evanescent piezoelectric field is weakened by the polarizability of water and the metal contacts on the Si samples. An electric field as high as possible is found to be advantageous for the deposition and alignment process of carbon nanotubes onto the target chip, since the strength of evanescent electric dipolar fields accompanying the surface acoustic wave and the micro-fluidic excitation of the suspension are key parameters. At significantly lower power levels of the launched surface acoustic wave, the success rate of aligning individual ropes of carbon nanotubes onto pre-structured contacts was considerably reduced. It has been shown in [17] by using a thin metal surface coating that the electric field accompanying the SAW is a dominant effect of CNT alignment, because there the electric SAW field is shorted and no alignment has been detected.

Typical results of deposition and alignment of SWNTs using SAW generated on a lithium niobate chip are depicted in the scanning electron microscopy images of figures 3(a) and (b), where individual SWNTs[2] bridges a gold contact pair separated by 500 nm to form a carbon nanotube junction. In figure 3(c), an individual MWNT (see footnote 2) is shown forming a carbon nanotube junction, while in the scanning electron microscopy image of figure 3(d), two individual MWNT form the carbon nanotube junction.

All carbon nanotubes were non-functionalized in order not to alter the original electronic properties, and merely adhesion forces are responsible for attaching carbon nanotubes to the contact pairs.

The deposition experiments described above were also performed using a lithium tantalate chip generating shear surface acoustic waves with a significantly lower potential to drive micro-fluidic flow fields. At the same power level of 5 mW, meaning approximately the same electric field but less streaming, the success rate in bridging gold contacts with carbon nanotubes using a lithium tantalate chip was only 10% compared to the $LiNbO_3$ case. As such, micro-fluidic agitation of the suspension-filled gap between the pre-structured silicon chip and the piezoelectric chip is an important element of the alignment process for the flip-chip devices. The second necessity is the electrical polarizability of carbon nanotubes along the tubular axis [22].

We have inspected all 90 contact pairs of the silicon target chips, and the statistics of the contacting process is shown in figure 4. It turned out that, whenever a contact pair was bridged by CNTs, most of the cases involved only a single CNT or two CNTs (see figure 4). For these first investigations of the

[2] Transmission electron microscopy investigations of the SDS-mediated carbon nanotube suspension used for deposition revealed that the individual SWNTs and MWNTs spanning a metal contact pair could also be ropes of SWNTs and MWNTs.

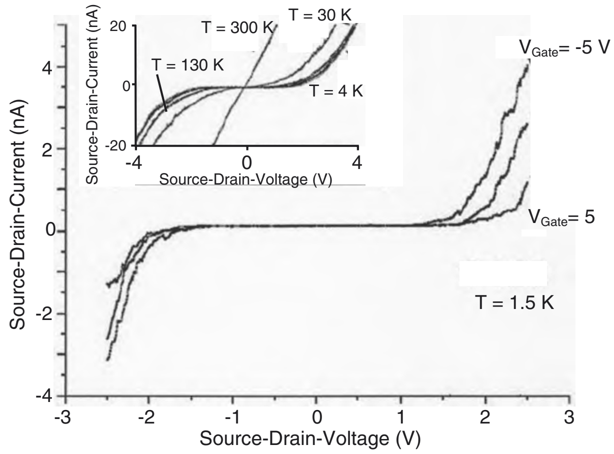

Figure 5. Electrical characterization of a CNT junction. Current–voltage measurements are presented for low temperatures together with their dependence on a back-gate voltage. The inset shows the current–voltage characteristic for different temperatures of the device without applying a back-gate voltage.

SAW-mediated CNT junctions we have achieved a bridging efficiency of 37% (33 contacts out of 90).

Electrical measurements of one of our typical prepared CNT junctions are shown in figure 5. For $T = 300$ K this device has an ohmic characteristic (see the inset of figure 5), but with decreasing temperature the device shows an increasing nonlinear behaviour. At a low temperature of $T = 1.5$ K, a pronounced nonlinearity of the current–voltage dependence has been measured. Apart from the non-perfect contacts between the source–drain metal and the CNT, a finite Schottky barrier and Coulomb blockade effects can be regarded as the origin for the non-ohmic behaviour at low temperatures. Applying a voltage to the silicon substrate back-gate changes the current–voltage characteristic of the device, indicating its possible application as a field-effect CNT junction.

4. Conclusion

In summary, we have demonstrated that SWNTs, as well as MWNTs, can be deposited and aligned between pre-structured metal contact pads of a silicon chip using surface acoustic waves. The lateral manipulation of carbon nanotubes is a major issue for CNT nanoelectronics as well as for fundamental research on the electronic and mechanical properties of carbon nanotubes.

Surface acoustic wave induced micro-fluidic flow fields assisted by electrical dipole fields within a thin film of aqueous carbon nanotube suspension are the driving forces in aligning carbon nanotubes.

Employing our alignment procedure, we have successfully fabricated a back-gated CNT junction of the tube-on-metal type. Electrical measurements have been performed with these devices, exhibiting a nonlinear current–voltage characteristic at low temperatures with an influence of the back-gate on device performance.

Additionally, our method for depositing and aligning carbon nanotubes is highly compatible with the separation of semiconducting and metallic SWNTs from an SDS-mediated aqueous suspension in a high-frequency field used in dielectrophoresis [20], making the on-chip fabrication of field-effect carbon nanotube junctions for fundamental research, as well as nanoelectronic applications, feasible.

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