High Frequency CNT Based Resonator for DNA Detection

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Abstract. The paper presents modeling and measurements of microwave propagation in CNTs based resonator for DNA detection. We report on sensing of adsorbed DNA on multi walled carbon nanotubes and deposited over a microwave IDT resonator. The DNA is revealed by a phase shift, of 20 degrees for the resonant frequency.

1. Introduction

The microwave devices based on carbon nanotubes (CNTs) constitute a promising area of research and many microwave devices, such as filters, resonators, amplifiers, and oscillators, which have been already built and tested in the frequency range of 0.5-100 GHz, and even beyond, up to THz. A recent review [1] gives a comprehensive overview of these efforts to bridge the nanotechnologies based on CNTs with microwave applications. The high impedance of CNTs make them very difficult to match CNTs-based RF devices or with any other RF device or circuit that have a standard impedance of 50 Ω. Recently, our group has obtained a resonator based on a CNT array in the microwave frequency range of 1–2 GHz. [2].

The detection of the deoxyribonucleic acid (DNA) represents a huge importance in molecular diagnosis of various diseases and early warning of serious illnesses. New DNA detection methods are required to be simple, fast and able to detect DNA with a reduced number of preliminary steps in the DNA sample processing. All these effective methods are termed free-label methods and carbon nanotubes (CNTs) play a leading role in them.[3] On the other hand, the dielectric properties of DNA are used to detect it at various electromagnetic frequencies.[4].

In this paper is reported the fabrication interdigitated structures IDT CNTs based resonators for DNA detection and also the experimental results at a high frequency.

2. Design and Modeling

The test structure was modeled and designed in frequency domain using CST Microwave Studio. CST uses time domain simulations and the port definition, S parameter extraction is optimized for electric field distribution (ideal for waveguide port and planar ports with multiple pins definition). The 3D view o resonator structure obtained by CST is presented in Fig. 1.

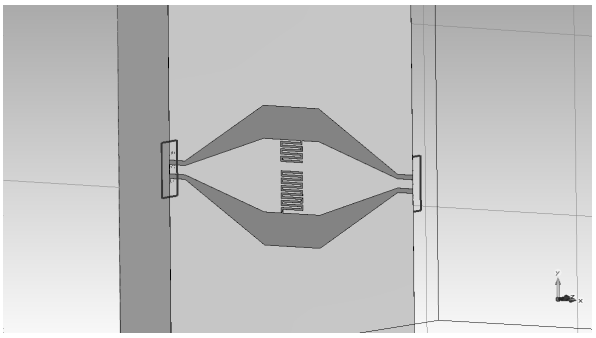


Fig. 3. D view of IDT resonator structure layout.

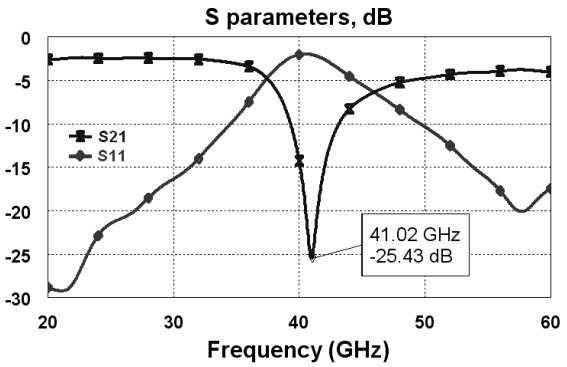


Fig. 2. Simulated S parameters for the IDT resonator.

The simulated S parameters are presented in Fig. 2. The simulated insertion losses are very good taking into account the large domain of frequency.

The design of the multilayer structure was made for a 10 (width)×140 (length) μm IDT structure having one large IDT for a 50 (width)×140(length) μm which transform a capacitor in a LC band stop resonator. The structure has been designed with the following layers:

- the substrate layer is Silicon, with high permittivity (11.9) and is 500 μm thick.
- the second layer is silicon dioxide (SiO_2), with a permittivity of 3.9 and 500 nm thick.

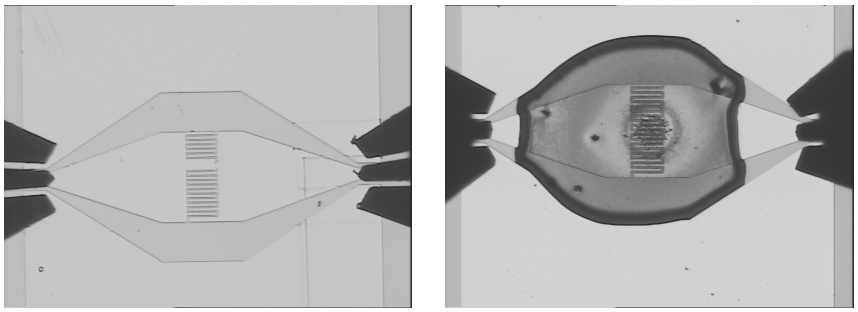
- the top layer is the metallization, where perfect electric conductor (PEC) was used, 500 nm thick.

The frequency bandwidth for this structure was 20-60 GHz and set in the environment. The next step was to set of some boundaries for the structure, so the open boundary was chosen for all the sides of the structure, and with space for the top side. In the final stage of the preparations for the simulation, the waveguide ports were designed, defining the coordinates for them, and selecting the type of the port, meaning Ground-Signal-Ground (GSG). The simulation was done using the Transient Solver, meaning time-domain.

3. Fabrication

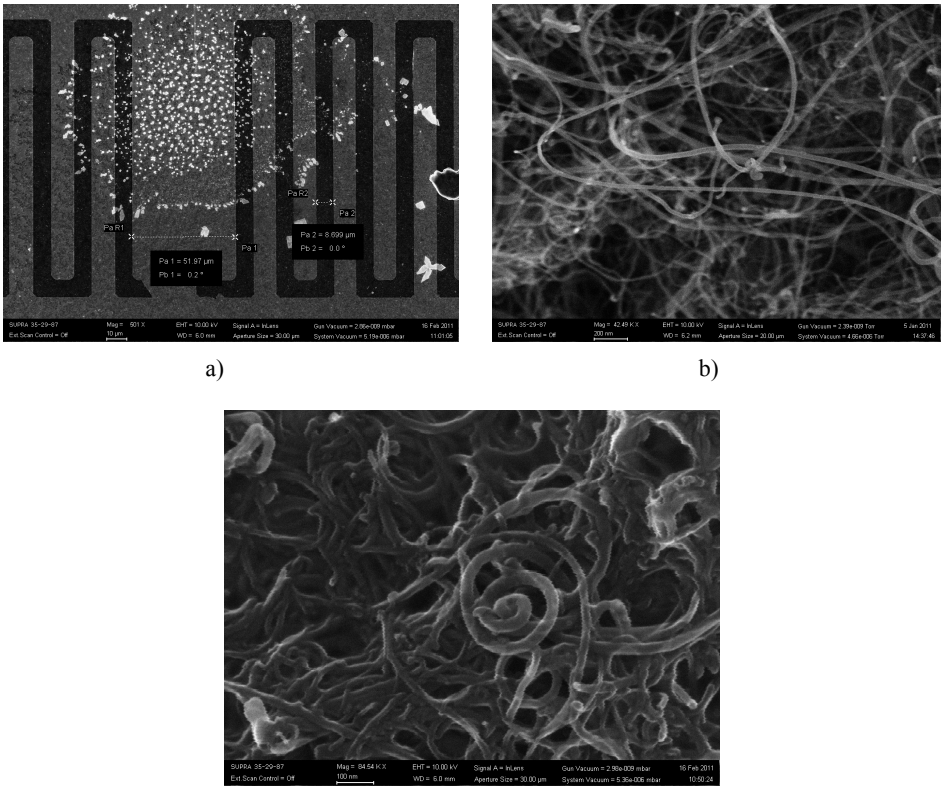
The IDT structure was manufactured on a SiO_2/high resistivity wafer with the dielectric permittivity 11.9. The fabrication has been done following the steps: (a) cleaning of the Si wafer (H_2SO_4 + H_2O_2, HF + DIH_2O), (b) thermal oxidation (t_{ox} = 1μm), (c) optical lithography consisting of treatment on hot plate at 110^0C for 10 minutes, AZ5214 photoresist deposition by spinning with thickness of 1.5 μm, and UV exposure and developing), (d) e-beam Ti/Au deposition with heat treatment during vacuum step, with layers of thicknesses 20 nm/80 nm, (e) metal lift-off (treatment on hot plate at 110^0C for 1 minute, till cracks appear on the metal surface, followed by cooling down to room temperature and then immersion in acetone, and (f) cleaning in isopropylic alcohol and DI water in a ultrasonic bath for 1 minute.

In the area of the IDT we deposited a drop (0.3 μL) of the MWCNTs and DNA composite with the following concentrations: 10 g /l CNT and 0.5 g/l DNA which can be seen in Fig. 3 (a) and (b) of the sensing device. The morphological characterisation of IDT resonator structure covered by CNTs and DNA has been obtained using scanning electron microscopy (SEM). As you can seen in Fig. 4 (b) and (c) texture differences are clearly visible i.e. before and after the DNA immobilisation.



a) b)

Fig. 3. Optical photo of the structure during the microwave characterisation.



a) b)

c)

Fig. 4. The SEM photo showing the deposition of the DNA- MWCNTs composite in the area of the IDT is displayed in Fig. 4 (a), while Fig. 4 (b) illustrates a detail of the CNTs before DNA immobilization, Fig. 4 (c) illustrates the detail of CNTs DNA composite after DNA immobilization.

The functionalization of MWCNT started with dispersing them for 6 hours, with the aid of ultrasounds, in 300 mL of concentrated H_2SO_4/HNO_3 (3:1, v/v) mixture. The suspension of purified CNTs was then diluted up to 1500 mL with distilled water and filtered on a 0.45 µm (Isopore™) membrane, using a Büchner funnel. The filtrate was washed plenty with distilled water until the pH of the waste water became 6.0. The functionalized nanotubes were afterwards dried in oven, at 100°C, for 4 h and the FT-IR spectra were acquired, showing the characteristic absorption band ascribed to the carboxyl group -COOH, situated at 1740 cm^{-1}. The interaction of λ-DNA (0.5 g/L) with MWCNTs-COOH (0.04 g/L), mixed at 1:1 v/v, was assayed spectrophotometrically (Fig. 5), using a photodiode array spectrophotometer (U-0080D Diode Array Bio-Spectrophotometer, Hitachi). At the

working pH, the adsorption process of λ-DNA on the MWCNTs-COOH backbone is much facilitated, being known that minimum influences on the adsorption processes concerning the immobilization of DNA on solid substrates are manifested in the 6.4 - 8.5 pH range when. We can see from Fig.5 that the composite DNA-functionalized MWCNTs displays a shape containing the spectral peaks of its constituents.

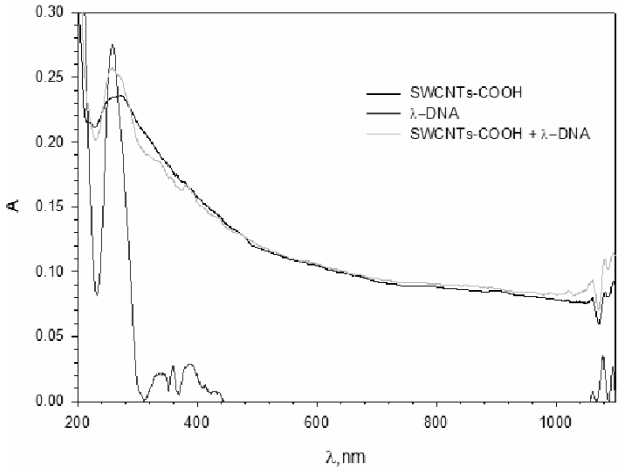


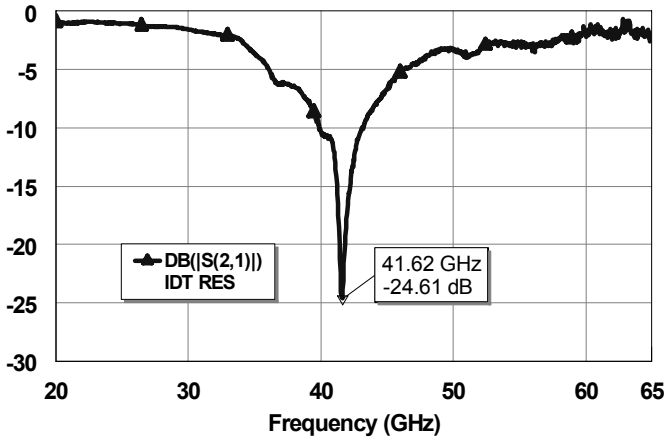
Fig. 5. UV-VIS spectra (in arbitrary units) of multi-*walled carbon nanotubes* (*MWCNTs,* black) and functionalized carbon nanotubes (MWCNTs-COOH, red) monitored spectrophotometrically (U-0080D Diode Array Bio-Spectrophotometer, Hitachi) is marked by the green color.

4. Measurement Results

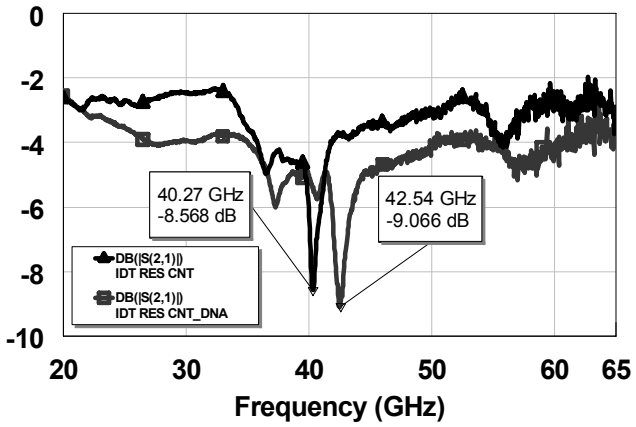
Further, we have performed the microwave measurements and for this purpose we have used two types of depositions made on the same wafer: IDT structure covered only with MWCNTs and IDT structure covered with the composite DNA - MWCNTs with the same concentrations as mention before. The two IDT structures covered with MWCNT and with DNA- MWCNTs functionalised were measured directly on-wafer with a vector network analyzer (VNA) -Anritsu - 37397D connected to a Karl-Suss PM5 on-wafer probe station. The SOLT calibration standard was used to calibrate the system.

The transmission of the IDT resonator structure is presented in Fig. 6 (a) and the results demonstrate a resonance of almost 41 GHz in a very good agreement with the simulation results. The transmission of IDT resonator structure covered with MWCNT and the DNA-MWCNTs composite are displayed in Fig. 6 (b).The results demonstrate a shift in frequency in comparison with the response of IDT

resonator structure respectively in the left side for the IDT resonator structure covered with MWCNTs and in the right side for the IDT covered with DNA-MWCNTs composite. We can see that the microwave signatures of the two compositions are quite different. The DNA signature is expressed by a distinct decrease of amplitude up to 20 GHz. Fig. 7 shows the corresponding phase for the two distinct depositions.



a)



b)

Fig. 6. S_{21} parameters for (a) IDT resonator structure, (b) IDT resonator structure covered with MWCNT and IDT resonator structure covered with DNA - MWCNT composite.

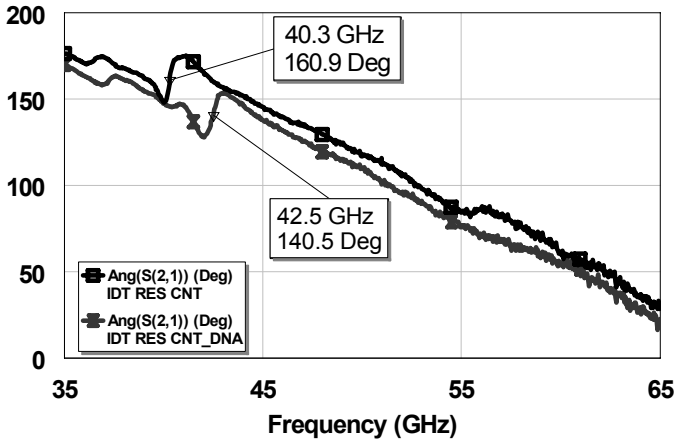


Fig. 7. The phase of the IDT resonator structure covered by MWCNTs and by the DNA- MWCNT composite.

The most interesting sensing parameter is the phase, which is shifted with 20 degrees in the range of resonance frequency for both structures (Fig. 7) and decrease at higher frequencies. The behavior in microwave range of MWCNTs and the DNA-MWCNTs composite IDT resonator structures can be explain by the differences in the microwave effective permittivity which is calculated as around 2 for MWCNTs and 11-58 for DNA-MWCNTs composite [4]. This is the reason why such a large phase shift occurs in Fig. 7 for the composite DNA-MWCNTs in contrast to the MWCNTs deposition. Repeated measurements over an interval of several days (not shown here) demonstrated that the electromagnetic responses of both the MWCNTs and the DNA-MWCNTs composite are stable in time.

5. Conclusion

In conclusion, we demonstrated using an IDT resonator structure covered by MWCNTs a detection method of DNA for a high frequency. The MWCNTs plays a key role in this sensitive detection procedure due to its low effective permittivity in microwaves, which contrasts the high permittivity of the DNA in the same frequency region. Instead of the decrease in quality factor of CNTs based resonator, the results are very important for the DNA sensing devices in high frequencies domain.

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