Remote Sensing Structures based on Surface Plasmon Resonances and Carbon Nanotubes

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***Abstract*— A sensor structure based on surface plasmon resonance (SPR) and carbon nanotubes (CNTs) is designed and developed. The device is an ultrasensitive wireless sensor operating in microwave frequencies utilizing the gas sensitivity of CNT mixtures. The sensor consists of a corrugated aluminum plate whose surface is periodically covered with a thin layer of the CNT materials. The incident TM-polarized waves on this surface excite the surface plasmon (SP) mode, thus resulting in a drop of power of the reflected wave. The simulations of the SPR-CNT-based wireless sensor show a frequency shift of 400 MHz in operation range around 22.5 GHz.**

I. INTRODUCTION

This paper introduces a practical application of surface plasmon resonance (SPR) for the realization of an ultrasensitive, fully-integrated, fully-packaged wireless sensor that can operate in microwave frequencies. In the current gas sensing technology, most gas sensor devices require some form of a direct physical contact. Carbon nanotubes (CNTs) have been found to have many distinct properties that may enable the next generation of sensors with a very high sensitivity up to 1 ppb (part per billion) [1-3]. Most CNT-based active sensors employ the change in resistance (easily affected by moisture), thermoelectric power, electrical breakdown voltage, or dielectric properties of the nanotubes for the gas detection [1]. Many of these sensors also require high temperature for operating conditions. Therefore a gas sensor that can operate at room temperature while maintaining a high sensitivity is highly desirable. Due to the strong demand for monitoring hazardous gases in remote or rugged environments, integration of the sensors with wireless monitoring technology is also desired for real-time remote sensing. The current method to utilize CNTs in any wireless system is based on the change in dielectric constant of the CNT composites/mixtures. While many types of direct physical contact sensors that utilize sensing properties of CNTs have been proposed, there have been very limited development on CNT-based wireless sensors due to the fact that the change of this parameter when the sensing CNT mixture is exposed to gas is around 1-4 % [2-4]. The difficulty in developing a remote sensing gas sensor is that the device needs to be based on the change in dielectric constant of the CNT mixtures which gives a very small variation when exposed to gas. Therefore an effective sensor needs to provide a mechanism that can magnify or project this small variation in the dielectric constant of the sensing material to an easier to observe or quantify effect. On the other hand, SPR is a charge-density oscillation that may be excited by incoming electromagnetic waves, and exists on the surface of two media with dielectric constants of opposite signs such as a metal and a dielectric. This charge oscillation is highly sensitive to dielectric properties of materials at the interface, therefore it may effectively transform the electrical change occurs on carbon nanotube materials induced by gas environment. Thus, we propose to use CNT mixtures/composites as the dielectric thin-film on an aluminum plate to realize a highly sensitive gas sensor that operates based on surface plasmons whose resonant frequency is determined by the air/aluminum/CNT dielectric interface.

II. OPERATION PRINCIPLES

SPR is a charge-density oscillation that may be excited by incoming electromagnetic waves, and exists on the surface of two media with dielectric constants of opposite signs such as a metal and a dielectric. SPR occurs under conditions of total internal reflection with p-polarized light (TM-polarized waves). The charge density wave is associated with the electromagnetic waves – the field vectors of which reach their maxima at the interface and decay evanescently into both media [5]. Since the propagation constant of surface plasmon waves (SPWs) in the metal layer is always higher than that of the wave propagation in the dielectric layer, the momentum of the incident wave has to be enhanced to match that of the SPW to achieve SPR. This is usually done by using total reflection in prism couplers or optical waveguides or by using diffraction at the surface of diffraction gratings where the periodic corrugation provides the necessary in-plane wave-vector enhancement in multiples of the grating vector. The working of the latter approach in coupling the incident wave to the SPW has been demonstrated for both optical and microwave incoming waves [6-7]. Since SPWs strongly depend on the interface conditions, their resonance condition is very sensitive to variations in the electrical properties of the dielectric adjacent to the metal layer that supports SPWs (noble metals, such as gold, silver, copper). The surface plasmon resonance (SPR) technique is an optical method for measuring the refractive index of very thin layers of material in contact with noble metals, which has enabled sensing for almost two decades. Since then, many sensors and biosensors based on optical methods, such as guided modes in optical waveguide structures (grating coupler, resonant mirror), and surface plasmon resonance have been developed [8-9].

In this work, we employ the grating technique to couple the microwave incident waves to SPs on the aluminum surface [6]. The wavevector kspr of an SPW along the interface can be written as

$$k_{spr} = \frac{\omega}{c}\sqrt{\frac{\varepsilon_1 \cdot \varepsilon_2}{\varepsilon_1 + \varepsilon_2}} \ , \qquad (1)$$

where ω is the radial frequency of the incident waves, c is the speed of light in vacuum, ε1 and ε2 are the complex permittivity of the two materials that form the interface (Fig. 1). However, the real part of the permittivity of aluminum has a large negative value in the microwave frequency range. Thus, Eq. (1) can be approximated as

$$k_{spr} = \frac{\omega}{c}\sqrt{\varepsilon_{CNT}} \ , \qquad (2)$$

where εCNT is the permittivity of the CNT composites/mixtures. The incident TM-polarized waves create an angle θ with the corrugated aluminum surface (Fig. 1). The resulting wavevector k_x on this surface can be written as

$$k_x = \frac{\omega}{c} \cdot \sin(\theta) + \frac{2\pi n}{\lambda_g} \ , \qquad (3)$$

where λg is the grating period and n is an integer (Fig. 2). The SPWs are excited under the condition where k_x is equal to k_{spr}. Therefore for a fixed incident wavelength λo with varying angle θ, the SPR yeilds a very sharp minimum at a particular angle θ in the reflected field; similar sharp minimum is observed when angle θ is fixed and λ_o is varying [9]. Using SPR transmission through a subwavelength hole or slit can also be enhanced significantly because the SPWs can be decayed by the grating and release radiated waves back into free space on the receiver side.

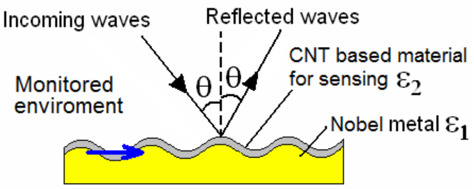

Fig. 1. Illustration of RF coupled into SPR.

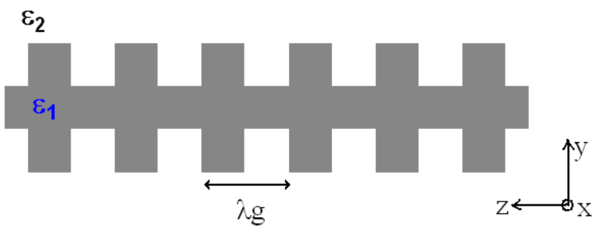

Fig. 1. Corrugated metal plate with ε1 adjacent to dielectric medium with ε2.

III. Modeling of the Carbon Nanotube Based Gas Sensing Materials in Simulations

Carbon nanotubes are typically manufactured through methods such as carbon arc-discharge technique, laser-ablation technique, or chemical vapor decomposition technique [10-12]. They can be single walled, double walled, or multi-walled and have a diameter typically around 1nm and a length typically around 1- 100 um. Their dielectric properties depend on the tube diameter, chirality, and doping [1]. The effective dielectric constant and attenuation constant of a double walled carbon nanotubes (DWNTs) mixture was investigated by Dragoman et al. in the frequency range up to 60 GHz [4]. It can be deduced from this work the dielectric constant and conductivity to be approximately 3.02 and 0.104 S/m respectively. The properties of the CNT composites can vary widely depending on the manufacturing methods, the weight percentage of the CNTs included in the mixtures and the operating frequency. The dielectric constant of the CNT composites strongly depends on the CNT weight percentage and has values in the range of 1-16 for an operating frequency range of 1-20GHz [13-14]. The conductivity of different mixtures of CNTs such as ceramic mixed with single walled nanotubes (SWNTs), epoxy mixed with SWNTs or multiwalled nanotubes (MWNTs), etc. were reported to have values in the range of 10-3 – 102 S/m [15]. The composites can also be modified to improve on the gas sensitivity by using functionalized CNTs [16].

At 10GHz, the electrical parameters of the DWNT are reported in [4] to have $\varepsilon_{eff} = 2.6$, and $\sigma = 1.57$ S/m where ε_{eff} is the effective dielectric constant, and σ is the conductivity. Based on the effective dielectric constant value and calculations performed in Agilent Design Systems (ADS), we deduce the dielectric constant of the DWNT mixture to be approximately 4.3. Our first objective is to observe the effect of small changes in dielectric constant on the SPR observed in a rectangular grating with a lossless dielectric on its surface. We consider the value of dielectric constant of DWNT at 10GHz and assume it to be lossless to allow for any changes in SPR to be solely due to the change in dielectric constant of the DWNT. Previously it has been shown that small changes in the dielectric constant of a multi-walled carbon nanotube (MWNT) - silicon dioxide (SiO_2) composite can produce small frequency shifts on a planar inductor-capacitor resonant circuit [2]. The MWNT-SiO2 composite was used as a superstrate. A change in the dielectric constant of the CNT base material of -0.09% for O_2 detection, -0.91% for CO_2 detection, and +2% for NH_3 detection was reported in [1]. The authors have chosen a dielectric constant change of +2% to compare the maximum measurable detection change at the maximum percentage change reported for the different approaches. All these changes in dielectric constant have been reported at approximately 18MHz. In [17], the difference in S21 magnitude and phase of a co-planar line used by Dragoman et. al. between reacted and non-reacted conditions becomes larger as the frequency is increased. The co-planar line has DWNT mixture as its substrate. Since the magnitude and phase of S21 depends on the dielectric constant and conductivity of the substrate, it evidently suggests that the response of the dielectric constant and conductivity of the DWNT mixture to gas increases as the operating frequency is increased. Therefore it is appropriate to use the change of 2% in dielectric constant observed in MHz range to implement in the dielectric model operating in the GHz range for testing the sensitivity of SPR structure in the worst case.

IV. DESIGN AND SIMULATION RESULTS

The detection performance using SPR for a small change of dielectric constant (2%) is investigated using the structure shown in Fig. 3, where the metal is aluminum and the CNT composite represents the dielectric. In this set up, h1 (13.9cm) is the distance from the transmitter waveport to the slit, and h2 (29.1cm) is the distance from the slit to the receiver waveport. The cross section of the structure is shown in Figs. 4a and 4b for the non-corrugated and corrugated samples. The dimensions of the structure are as follows: a = 2mm, b = 4mm, t = g = h = 4mm, and λg = 4mm. In this set up $\varepsilon r'$ is set to 4.30 in the absence of the gas, and $\varepsilon r'$ is set to 4.386 in the presence of the gas; $\varepsilon r''$ is set to zero for lossless assumption. The simulation is performed in FDTD Solutions (evaluation version) by Lumerical Solutions, Inc. TM-polarized waves are incident on the corrugated metal plate covered with a CNT material layer of thickness t. The transmission of this power through the subwavelength slit is collected by a receiver on the other side of the metal plate as shown in Fig. 3. Simulation results are shown in Fig. 5 for three cases: aluminum plate covered by dielectric layer with no grating in the absence of gas, aluminum plate covered by dielectric layer with grating in the absence of gas, and aluminum plate covered by dielectric layer with grating in the presence of gas. The results show a sharp peak of enhancement around 22 GHz in comparison to the non-corrugated structure (case a in Fig. 5) and the corrugated one in the absence of gas (case b in Fig. 5). In the absence of gas, the enhanced peak is at 22.6 GHz with a magnitude of 0.218. In the presence of gas, this peak is shifted to 22.2 GHz with a magnitude of 0.208. The frequency shift is about 400 MHz. This result successfully demonstrates our proof-of-concept on the use of SPR for gas sensing with extremely small change (2%) in the dielectric constant of the sensing material.

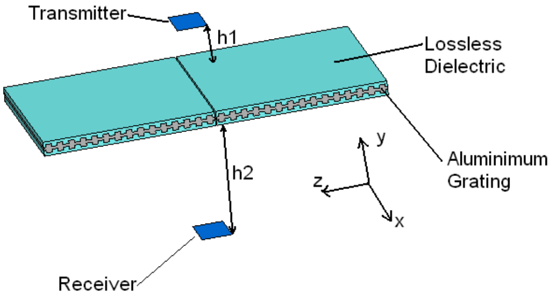

Fig. 3. Simulation set up for sensing using the enhancement of SPR in transmission with 2% change in the sensing dielectric represents CNT composite

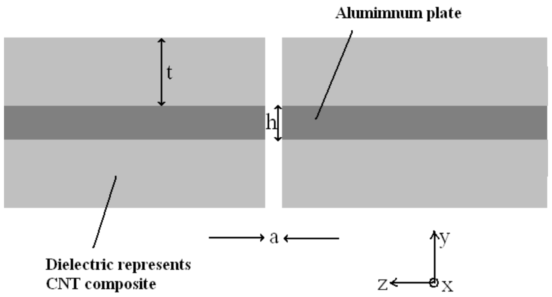

Fig. 4a. Cross section of the slotted sample without grating in transmission set up

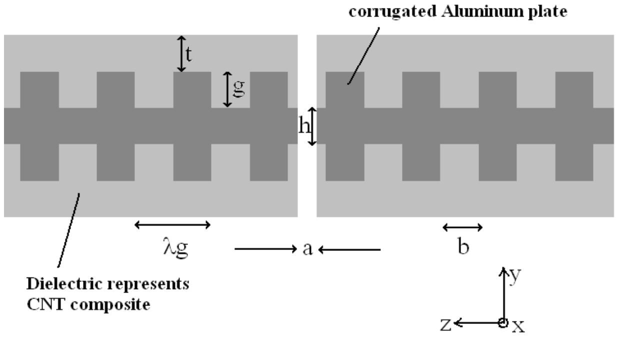

Fig. 4b. Cross section of the slotted sample with grating in transmission set up

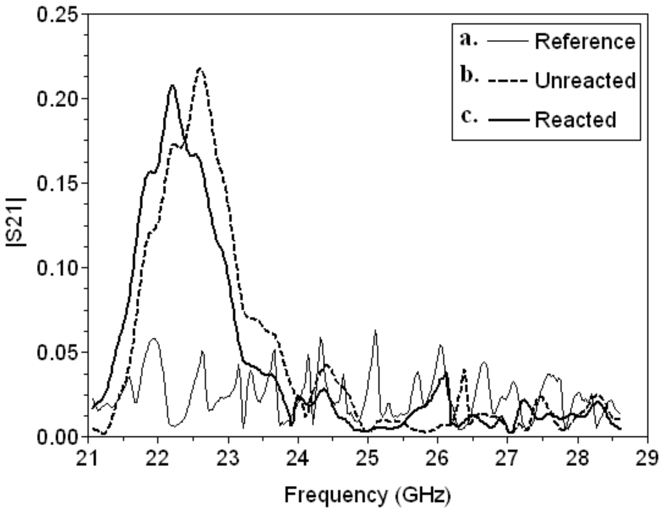

Fig. 5. Magnitude of the transmission through the subwavelength slit plotted against frequency for a) the reference simulation of non-corrugated plate covered by unreacted dielectric layer, b) the simulation of corrugated plate covered with un-reacted dielectric layer, and c) the simulation of corrugated plate covered with reacted dielectric layer.

V. CONCLUSIONS

We have presented a novel design for sensing gas based on in Surface Plasmon Resonances and Carbon Nanotubes that operate in microwave frequencies. This same SPR design can also be implemented in other type of sensing that is based on the electrical change of the sensing materials. The sensor presented in this paper is passive and ultrasensitive and miniaturized, giving a shift of 400 MHz. Based on the shift in SPR and the level of reflectivity, potential selectivity in sensing may also be achieved. The SPR approach also allows further miniaturization as the size of the sensor is decreased when the operating frequency is increased without affecting the sensitivity of the sensor. Although the recovery time of the CNT composites is slow (6-10 hrs) which is the limitation of all gas sensors based on CNTs, the response time is very fast within a few minutes. This feature together with high sensitivity and remote sensing capability may allow detection of cracking in spacecraft in real-time (based on detection of gas leakage through the cracks), or monitoring the leakage of dangerous gases into the public and private environments.

ACKNOWLEDGMENT

The authors are thankful to the supports of NSF ECS-0801798, IFC, and GEDC.

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