

Shot Noise Thermometry for Thermal Characterization of Templated Carbon Nanotubes

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Abstract—A carbon nanotube (CNT) thermometer that operates on the principles of electrical shot noise is reported. Shot noise thermometry is a self-calibrating measurement technique that relates statistical fluctuations in dc current across a device to temperature. A structure consisting of vertical, top, and bottom-contacted single-walled carbon nanotubes in a porous anodic alumina template was fabricated and used to measure shot noise. Frequencies between 60 and 100 kHz were observed to preclude significant influence from $1/f$ noise, which does not contain thermally relevant information. Because isothermal models do not accurately reproduce the observed noise trends, a self-heating shot noise model has been developed and applied to experimental data to determine the thermal resistance of a CNT device consisting of an array of vertical single-walled CNTs supported in a porous anodic alumina template. The thermal surface resistance at the nanotube–dielectric interface is found to be 1.5×10^8 K/W, which is consistent with measurements by other techniques.

Index Terms—Carbon nanotubes, electrical noise, thermal resistance.

I. INTRODUCTION

CONTINUED reduction in the size of engineered micro and nano-scale devices has created a critical need for improved thermal transducers to characterize effects such as heat conduction mechanisms, electrical self-heating, and phase change phenomena at ultrasmall scales. An ideal transducer would be spatially compact, be self-calibrating, and allow non-invasive *in situ* measurements. Further, because of their ultrasmall sizes, such transducers could be designed in integrated

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platforms that would bring unprecedented spatial, temporal, and statistical resolution.

Today, most solid-state thermometers for small-scale measurements employ a technique that relates changes in electrical resistance to temperature. Devices used in such resistance thermometry must have a resistance–temperature curve that is calibrated with high accuracy and precision. Also, the device's resistance must remain stable over time and operating conditions to prevent measurement drift and frequent recalibration. Satisfying these conditions is particularly challenging in nanoscale structures such as nanowires and nanotubes. For example, small changes in synthesis conditions can lead to large changes in the resistance of multi-walled carbon nanotubes (MWCNTs) [1]. Further, the resistance of single-walled carbon nanotube (SWCNT) devices has been shown to vary dramatically with contact type, length, and conducting type [2]–[5] and such variance greatly limits the applicability of resistance thermometry in these devices. In addition, the small temperature coefficient of resistance of carbon nanotubes (CNTs) (typically one to two orders of magnitude lower than typical metals used in resistance thermometry such as Pt [6]) reduces transducer resolution. In addition, other researchers have found that CNTs have a negative temperature coefficient of resistance [7]–[9], which requires modification of typical resistance thermometry.

An alternative approach to temperature measurements in CNTs involves the fabrication of a structure that resembles a common mercury thermometer. The first such thermometer was reported by Gao and Bando [10] and involved filling the hollow CNT core with gallium. Although an intriguing development, widespread application of this type of CNT thermometer is limited owing to the fact that the Ga level must be monitored by an electron microscope. Liu *et al.* [11] simplified the previous technique by eliminating the initial calibration step and decreased the error of measurement below 5%. Dorozhkin *et al.* [12] utilized a two-order of magnitude difference in the resistivity of a Ga-filled portion of a CNT compared to an empty CNT to relate the electrical resistance of a Ga-filled CNT to its temperature. This method allows real-time measurement of temperature changes. However, calibration of the thermometer is very complicated and time-consuming, and the temperature-dependent electrical resistivity of the Ga-filled CNT and empty CNT must be measured using an atomic force microscope.

Although the foregoing developments in nanoscale thermometers are highly innovative, all such thermometers require calibration that can be complicated and sample-specific. Noise thermometry offers a compelling alternative to resistance thermometry for nanoscale objects. A shot noise thermometer made from a CNT would act as a self-calibrating transducer that accommodates variations in electrical behavior that are difficult and sometimes impossible to control precisely.

Sensors used to measure temperature can be divided into two groups: primary and secondary thermometers. Primary thermometers are characterized by well established state equations that relate the measured quantity to temperature through a simple physical principle. These thermometers are capable of providing absolute temperature without calibration, while secondary thermometers, such as those that exploit changes in electrical resistance, must be calibrated to a known temperature. Most primary thermometers are expensive and difficult to operate; however, electrical noise measurements can provide a simple basis for creating primary thermometers that are capable of accurate temperature measurements in the range from 1 mK to greater than 1500 K [13].

This paper presents results from a carbon nanotube sensor that utilizes a unique combination of innovative nanofabrication processes and a primary temperature measurement approach that exploits nanoscale transport principles. Electrical noise (Johnson and shot noise) measurements of SWCNTs in a porous anodic alumina (PAA) template in combination with a self-heating model are used to determine thermal resistance of the SWCNTs.

II. THEORY

A. Johnson Noise

The electrons in electrical conductors are free to move as a so-called electron gas. Even in the absence of an applied bias, statistical fluctuations of the electric charge in the conductor occur due to the random movement, or Brownian motion, of electrons caused by thermal agitation. This effect was first observed and measured by Johnson [14], [15] for various conductors using a vacuum tube amplifier and was explained theoretically by his colleague, Nyquist [16], using the equipartition theorem. This effect of thermal noise garners its name from the early pioneers of the field as Johnson–Nyquist or simply Johnson noise.

From quantum theory, the average energy W associated with each oscillating mode of frequency f is given by

$$W = \frac{hf}{\exp\left(\frac{hf}{k_B T}\right) - 1} \quad (1)$$

where h is Planck’s constant, k_B is Boltzmann’s constant, and T is absolute temperature. If $hf \ll k_B T$, (1) reduces to $W \approx k_B T$. The voltage spectral density S_V and current spectral density S_I are expressed as

$$\begin{aligned} S_{V,J} &= 4k_B TR \\ S_{I,J} &= \frac{4k_B T}{R} \end{aligned} \quad (2)$$

where R is the resistance of the conductor which may vary with frequency and the subscript J denotes Johnson noise.

Equation (2) is independent of frequency and is valid for frequencies up to several hundred GHz at room temperature [17].

B. Shot Noise

Shot noise, first reported by Schottky [18], occurs in any system in which discrete charge carriers move across a potential barrier, such as in cold cathodes, tunnel junctions, and PN junction diodes. Electron transport through a tunnel junction is statistical in nature and depends on the size of the barrier, as well as the potential between the anode and cathode [19]. Although the current through the junction averaged over a sufficiently long time is constant under a steady applied bias, the rate of electrons passing through the junction fluctuates randomly at short time scales. Each electron that tunnels through a barrier can be considered as a discrete “shot” or impulse of charge and can be represented by a Dirac delta function. The current spectral density of shot noise is [20]

$$S_{I,s} = 2eI \quad (3)$$

where e is the electron charge, I is the average current, and the subscript s denotes shot noise. Like Johnson noise, shot noise is independent of frequency up to several hundred GHz.

Because shot noise is independent of temperature, it may appear to be of little use in temperature measurements. However, when measured in combination with Johnson noise, Spietz *et al.* [21], [22] showed recently that shot noise can be exploited to achieve accurate temperature measurements from the mK range to many hundreds of Kelvin. The current spectral density for combined Johnson and shot noise is given as [23]

$$S_{I,J+s} = 2eI \coth\left(\frac{eV}{2k_B T}\right). \quad (4)$$

At zero applied bias this equation reduces to the Johnson form, $S_{I,J} = 4k_B TR$ and at large applied biases it approaches the shot noise limit, $S_{I,s} = 2eI$.

III. ANALYSIS

A. Self-Heating Model

The prior discussion assumes that the temperature of the device under test remains constant with applied voltage. In practice, the temperature of the device varies with bias due to joule heating. Accounting for self-heating, (4) can be written as

$$S_{I,J+s} = 2eI \coth\left(\frac{eV}{2k_B T(V)}\right) \quad (5)$$

where $T(V)$ is the voltage-dependent temperature. By assuming electron–phonon equilibrium and diffusive heat and current flow in the SWCNT, $T(V)$ is given by

$$T(V) = QR_\theta + T_\infty \quad (6)$$

where $Q = IV = V^2/R$ is the heating rate, T_∞ is the ambient temperature, and R and R_θ are the electrical and thermal resistances, respectively. Because of the short decay length of heat from the contact into the CNT, the voltage-dependent temperature of (6) is assumed to be independent of

position along the CNT. In SWCNTs, significant self-heating occurs when the power density is greater than $5 \mu\text{W}/\mu\text{m}$ [24]. Substituting (6) into (5), the temperature-dependent shot noise relation is

$$S_{I,J+s} = 2eI \coth \left(\frac{eV}{2k_B(QR_\theta + T_\infty)} \right). \quad (7)$$

The thermal resistance between a CNT device and its surroundings will likely be dominated by the nanotube–substrate interface [25]. In fact, a similar study has reported that approximately 80% of Joule heat generated in a SWCNT is dissipated to the surrounding medium and only 20% to the metallic contacts [26]. This thermal surface resistance can play a major role in limiting heat flow from the CNT to the surrounding medium [27], [28], which can lead to large temperature rises. Shi *et al.* [29] measured the thermal resistance of suspended SWCNT bundles using a microfabricated device to be $\approx 4 \times 10^8$ K/W and $\approx 5 \times 10^7$ K/W and for bundles of diameter 10 nm and 148 nm, respectively. Dames *et al.* [30] used a hot wire technique to measure the thermal resistance of individual MWCNTs contacted with liquid Ga inside a transmission electron microscope. They reported a value of 3.3×10^7 K/W for the combined thermal resistance of the CNT and contacts.

IV. EXPERIMENTAL SETUP

A. Device Fabrication

The CNT thermometer developed in this paper is composed of an array of 400 nm long vertical SWCNTs grown in a PAA template with an embedded Fe catalyst layer [Fig. 1(a)] according to a process recently reported [5], [31]–[35]. Carbon nanotubes were grown within nanoscale pores by a microwave plasma chemical vapor deposition process. The CNT synthesis was conducted at a substrate temperature of 900 °C with gas flow rates of 50 sccm H_2 and 10 sccm CH_4 and a plasma power of 300 W at a pressure of 1.3 kPa.

Subsequently, Pd nanowires were electrodeposited into the pore bottoms [Fig. 1(b)] to make an electrical contact to the CNTs [31]. Top contacts [Fig. 1(c)] of Pd/Ti/Au were defined using photolithography and deposited via electron-beam evaporation. The final device was inspected with a cold field emission scanning electron microscope (FESEM). A tilted view of the top contact is shown in Fig. 1(d). The top contact was evaporated onto the CNT covered PAA surface. The CNTs appear as bright lines on the top alumina surface due to electron beam charging effects [32]. The inset shows a cross sectional view prepared by fracture of the structure.

Room temperature I – V characteristics of the device are shown in Fig. 2. The linear relationship between voltage and current indicates ohmic contacts to the CNTs. The overall device resistance was 359 Ω . Franklin *et al.* [35] measured the electrical resistance of an individual metallic SWCNT grown from PAA with an estimated length of 100 nm to be ~ 125 k Ω . In [35], the portions of the SWCNTs extending along the PAA surface were removed with an etching process that left only the tips of the SWCNTs exposed for contact. The device reported here has SWCNTs that protrude from the pores of

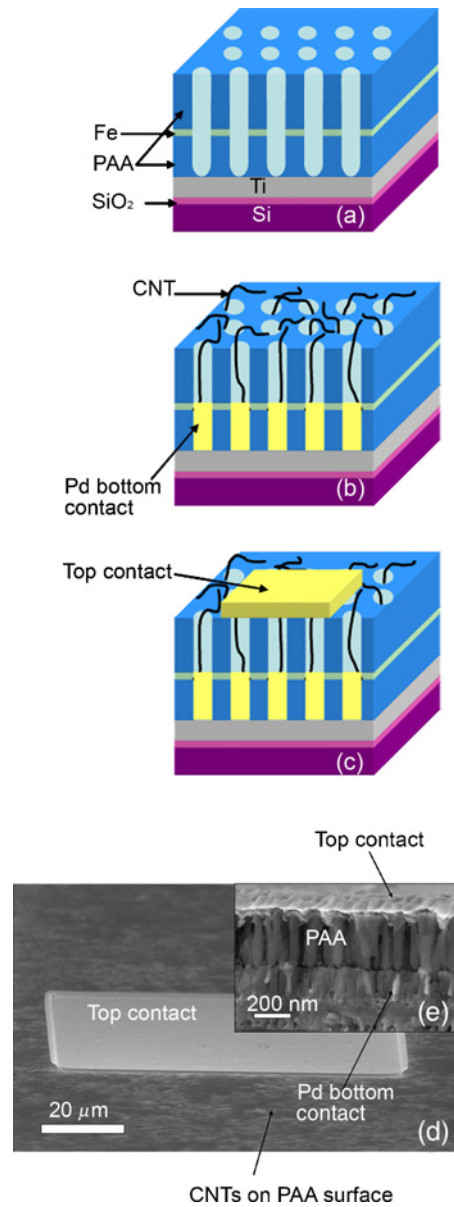


Fig. 1. Fabrication procedure for the CNT shot noise thermometer. (a) PAA with an embedded Fe layer is used as a template. (b) SWCNTs are grown at a density of no more than one per pore and Pd nanowires are deposited. The length of the SWCNTs in the PAA template is 400 nm. (c) A top contact is evaporated to the CNT tips. (d) Tilted FESEM image of the final device. The $80 \mu\text{m} \times 80 \mu\text{m}$ top contact pad is shown on top of the CNT covered PAA surface. CNTs appear as bright lines covering the PAA surface. (e) Cross-sectional view of the final device.

the PAA and extend along the surface, thus allowing for edge and tip contact of the SWCNTs and reducing the resistance between the SWCNTs and the top contact. Therefore, 125 k Ω is taken as an upper bound of the resistance of an individual CNT, although the actual resistance should be much less due to the improved contact. Furthermore, Franklin *et al.* [35] showed that semiconducting SWCNTs in a PAA template have a resistance of approximately 50 M Ω because they are electrically turned off. Assuming that all of the SWCNTs in the test device are connected electrically in parallel, we estimate the number of contacted tubes to be no more than 350. Further, all low-bias electrical transport is assumed to be

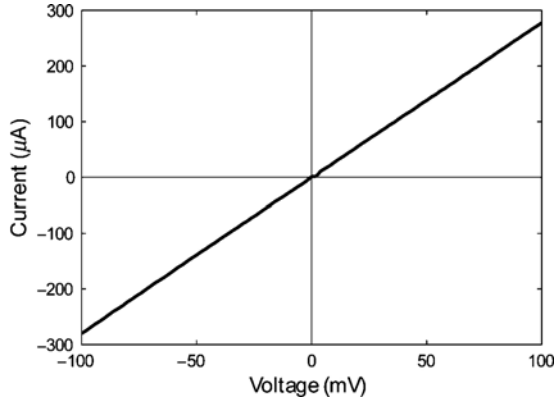


Fig. 2. Room temperature I - V characteristics of a typical vertical SWCNT device.

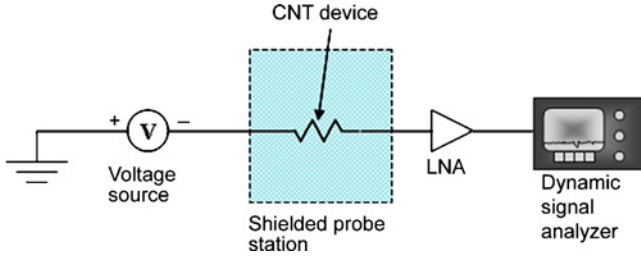


Fig. 3. Schematic of the setup used to measure electrical noise. The dynamic signal analyzer is rated for measurements from dc to 102.4 kHz and the LNA had a gain $200 \mu\text{A}/\text{V}$.

through metallic SWCNTs, although the specific chirality of the SWCNTs is not known.

B. Measurement System

A constant dc voltage is applied across a SWCNT device that is placed inside a shielded probe station in order to minimize external noise injection. The noise across the device is then amplified with a low noise current preamplifier (LNA) and measured with a dynamic signal analyzer as shown in Fig. 3.

V. RESULTS

DC voltage biases ranging from 0 to 80 mV were applied to the CNT array in a room temperature (296 K) ambient within a shielded probe station. The current spectral density from 3 to 100 kHz was measured. 150 root mean square (RMS) data averages were computed to minimize statistical fluctuations in the measured noise spectra. The current spectral density of noise is shown in Fig. 4 for several different voltages. Instrument noise (resulting from both the dynamic signal analyzer and LNA) was measured and found to be constant with dc bias. The value of instrument noise floor (about $10^{-21} \text{A}^2/\text{Hz}$) was subtracted from the subsequent measurements.

At frequencies below 60 kHz, the spectra are dominated by $1/f$ noise, consistent with previous studies [36]–[38]. Also, the slight divergence of the noise levels approaching the highest frequency (10^5 Hz) is a result of reaching the frequency limit of the dynamic signal analyzer. At intermediate frequencies,

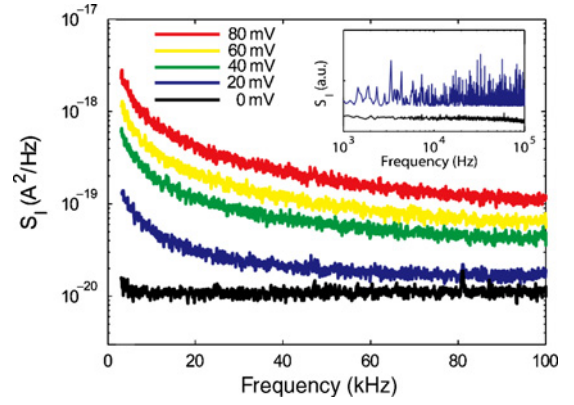


Fig. 4. Current spectral density measured across the CNT device. $1/f$ noise dominates the signal at frequencies below 60 kHz. Shot noise is observed at frequencies above 60 kHz. Inset: Measured current spectral density at 0 mV using 1 (top curve) and 200 (bottom curve) RMS data averages.

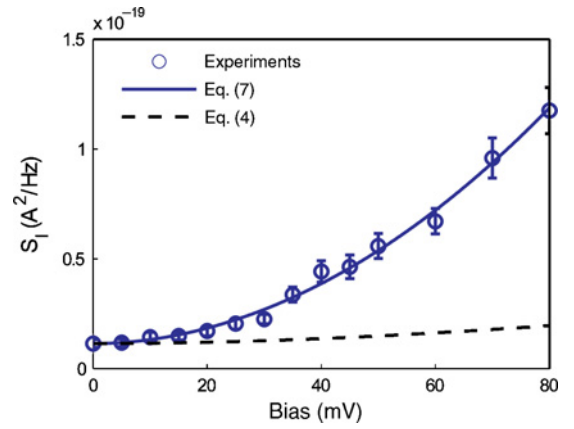


Fig. 5. Experimentally measured shot noise current spectral density as a function of dc bias voltage. The dashed line is the value predicted without self-heating and the solid line is a fit to the data including self-heating. $R_\theta = 1.5 \times 10^8 \text{ K}/\text{W}$. The error bars represent one standard deviation from the mean upon averaging over the frequency independent portion of the spectrum.

the measured noise is independent of frequency, but its average magnitude varies with applied dc bias as is characteristic of shot noise. The inset of Fig. 4 shows the measured noise signal at 0 mV with 1 and 200 averages. The curves are offset vertically to enable visual distinction between the two. Although both measurements fluctuate about the same mean value, averaging reduces the amplitude of these fluctuations.

The average current spectral density (averaged over the frequency-independent portion of the spectrum) is shown in Fig. 5 for dc biases ranging from 0 to 80 mV. The error bars in the figure represent one standard deviation from the mean value averaged over the range 79–100 kHz. Notably, the self-heating model (7) accurately predicts the shape of the measured shot noise curve, whereas an isothermal model (4) fails to capture the trend in noise with increasing voltage bias. A fit of the average noise data to (7) yields a thermal resistance of $1.5 \times 10^8 \pm 0.1 \times 10^8 \text{ K}/\text{W}$, where the uncertainty is based on the 95% confidence interval of the curve fit. The order of magnitude of the resulting thermal conductance per unit CNT length (with a 400 nm array height) is therefore $g \sim 0.1 \text{ W}/\text{Km}$. Notably, this result agrees well with a recent

estimate by Pop *et al.* [24] of $g = 0.17$ W/Km for overall thermal conductance for a self-heated metallic SWCNT on a dielectric substrate based on a combination of experimental measurements and coupled electro-thermal transport models.

The temperature rise in the CNT device can be calculated from (6). For example, when a current of $100 \mu\text{A}$ is applied to the device, the estimated temperature rise is $\approx 540^\circ\text{C}$. While this projection seems unexpectedly high, we note that current will almost certainly channel through the least resistive pathways in the massively parallel array, and therefore, heat dissipation is expected to be localized to a small fraction of the total number of CNTs. The reported temperature rise is that experienced by the group of most-heated CNTs in the array. Temperature rises of this magnitude have been previously observed in individual MWCNTs suspended in a vacuum where breakdown temperatures are reported to be in the range of $1800\text{--}3000^\circ\text{C}$ [39], [40]. Assuming the breakdown temperature to fall in this range and the breakdown current of a SWCNT to be $18\text{--}25 \mu\text{A}$ [41]–[43], we approximate the resistance of an individual SWCNT to be $23\text{--}67$ k Ω and the actual number of contacted CNTs to be between 64 and 186. Further work is ongoing to isolate such effects by studying individual CNT devices and small areas of the structure discussed above.

VI. CONCLUSION

Shot noise of a device consisting of a parallel array of vertical SWCNTs supported in a PAA template has been measured. The magnitude of the shot noise does not follow the expected curve for an isothermal device. Rather, a substantial deviation due to significant self-heating of the CNT device is observed. A simple self-heating shot noise model that assumes constant device resistance has been developed and provides a good fit to the measured data. The resulting thermal resistance of the CNT device is found to be $1.5 \times 10^8 \pm 0.1 \times 10^8$ K/W. This technique offers promise for further development for the study of individually contacted single and multi-walled CNTs to investigate electrical self heating in CNT transistors and the Nottingham effect in field emitters.

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