Diagnosing Nanoelectronic Components Using Coherent Electrons

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Supporting Information

ABSTRACT: We present the direct observation, using off-axis electron holography (EH), of the electric potential distribution in the vicinity of a single carbon nanotube electrically biased by two closely spaced contacts. When our results are combined with finite element modeling, we demonstrate the ability to separately observe the electrostatic potential drops across the metal contacts at the interface with the nanotube and along the length of the nanotube itself. We demonstrate that the uneven resistivity of different contacts can cause an asymmetric EH phase shift, which can readily be identified and quantified. EH thus offers a unique and precise approach for in-depth understanding and quick diagnosis of many similar nanoscale electronic devices.

KEYWORDS: Carbon nanotube, electron holography, electric potential, contact resistance, device diagnosis, diffusive transport

Advances in nanotechnology have enabled many new forms of nanoelectronic components implemented using emerging nanomaterials and nanostructures, such as nanotubes, nanowires, graphene, organic thin films, and single molecules.1−4 With the increasing demand for size reduction and integration, reliable nanofabrication and precise device characterization are crucial to push applications forward. To truly understand how a functional component works and why a defective part misbehaves on the nanoscale, many characterization techniques are typically involved. For the particular interest of detecting failures in nanoelectronic devices, conventional transport measurements give limited information and are blind to direct spatial details at the nanometer-scale. Electron-beam-induced current imaging, based on scanning electron microscopy, has been widely used to identify failure locations and p−n interfaces in semiconducting devices, but it cannot reveal quantitative information about electrostatic potentials.5,6 Scanning probe techniques, such as scanning gate microscopy and Kelvin probe force microscopy,7,8 can obtain localized signals of surface electric potential by locally recording electrostatic tip−sample interactions, but the measurements are susceptible to surface artifacts and unwanted dipole moments due to work function variations.9 On the other hand, electron holography (EH), which utilizes coherent electrons in a transmission electron microscope (TEM), can directly record the electrostatic potential without such drawbacks. It has been employed for detecting magnetic induction fields,10 determining a semiconductor’s doping levels and junction barriers,11 and mapping the electrostatic potential within as-fabricated nanoscale transistor devices.12,13 EH has also been used to study the electronic properties of carbon nanotubes, such as measuring the material-intrinsic mean inner potential14,15 and observing the electric-field distribution around a carbon nanotube with a single electrical contact.16,17

Here we show that EH may also be used to study a carbon nanotube with two electrical contacts and at much lower biases, allowing the determination of the low-voltage contact resistance of such a nanoelectronic device, separately from its intrinsic internal resistivity.

Carbon nanotubes, as a symbolic one-dimensional material with outstanding electrical, optical, thermal, and mechanical properties, have been utilized as a core component of nanoelectronics, nanophotonics, and optoelectronics,18,19 as well as in new applications in emerging energy technologies such as photovoltaic and rechargeable battery devices.20,21 Among all these applications, the configuration of a carbon nanotube (CNT) connecting with a source and a drain is considered as a fundamental prototype structure to function in a complex nanosystem, such as a field-effect transistor.22,23 The electronic transport properties of a single CNT have been experimentally explored extensively using DC electrical measurements with many results showing diffusive24−26 as well as ballistic24−26 transport under various conditions and device histories. In this Letter, we utilize a single-CNT device as an idealized model of a nanoelectronic device to address these questions of fundamental concern, through direct quantitative imaging of the electric potential in the vicinity of the CNT and its contacts.

Figure 1a illustrates the configuration of a single-CNT device and the principle of EH. The CNTs in this study were synthesized by chemical vapor deposition and are multiwalled nanotubes (MWNTs) with diameters around 50 nm and lengths up to a few micrometers. The prototype device chips were customized from commercial 50 nm Si3N4 membranes on
Si TEM grids. First, Au electrodes were patterned onto the nitride membrane using electron beam lithography and a lift-off process to make electrical contact with a homemade TEM holder capable of electrical biasing and measurement. Then, multiple arrays of open slits, each around 1 μm wide and 10 μm long, were fabricated by fluorine reactive ion etching through a polymer mask, also patterned by electron beam lithography. The CNTs were dispersed by sonication in isopropanol and then spin-deposited onto the TEM chip. After locating proper individual CNTs that conveniently cross a slit, another layer of 100 nm Pd was lithographically patterned to make electrical connections from the Au electrodes to the CNTs. The TEM image of an actual single-CNT device is depicted in Figure 1b. The extra etching step to remove the nitride membrane underneath the CNT enables a more precise phase retrieval by the zero-bias phase shift from the other phase-shift images, we can determine the MIP for this CNT to be 10.9 V, which is in agreement with previous reports. By subtracting the zero-bias phase shift from the other phase-shift images, we can remove the MIP and any other systematic background artifacts and field perturbations caused by localized charging effect and/or shape variation of the electrodes and thus obtain the pure phase changes arising from the CNT potential. Figure 2a shows the processed phase images at applied voltages of ±0.2, ±0.4, ±0.6, ±0.8, and ±1.0 V. These phase shifts appear roughly in the same asymmetric oval-like shapes with only differences in the magnitude, as measured by the number of color contours. To quantify the phase change, a line profile across the CNT position from the phase image obtained with ±1.0 V bias is plotted in Figure 2b, in which a downward phase slope is measured to be –2.86 rad/μm. Similar analysis is performed at the exact same position of the other phase images, and an obvious linear dependence between phase gradient and applied voltage is demonstrated in Figure 2c. It is in fact physically meaningful that the projected electric field is proportional to the voltage, so that acquiring the measurable phase shift can provide direct quantification of the voltage applied on the device. It is reasonable to expect that nonlinear behavior arising from a rectifying device can also be quantified.

We point out a peculiarity of our measurement technique that dictates that a conventional diffusive conducting CNT with low-resistance (LoR) contacts should give no measurable signal in our phase-shift images, other than the MIP signal. The reason for this is that the potential drop between the Pd electrodes may be considered as a capacitor, with a transversely
uniform and laterally linear change in potential from one electrode to the other (the transverse uniformity is generically justified, but the lateral linearity is only strictly valid for parallel-planar electrodes, see Supporting Information). The potential in a uniform diffusive conductor should exhibit the same linear potential change due to the internal electric field and thus the difference in potential, as is recorded in our phase-shift maps, should be negligible under such conditions. Measurable phase shifts can be produced by deviations from these conditions, for example, by a high interface resistance (HiR) at one or both electrical contacts. As we show below, these phase shifts allow us to directly determine which contacts exhibit excess resistance and to quantitatively assign an amount of resistance to the contact and an amount of resistance to the bulk of the CNT itself.

For a better understanding of the phase distribution, finite element modeling has been performed using the COMSOL Multiphysics software. The strategy for these calculations is illustrated in Figure 3. In a generic situation considering all the electrostatic aspects, if we apply voltage $V_{E1}$ and $V_{E2}$ on the top and bottom electrodes, respectively, the actual voltages on both ends of the CNT are $V_{C1}$ and $V_{C2}$, respectively, due to contact resistance, as shown in region (i) of Figure 3a. However, in the reference area (denoted as region (ii)), a linear potential profile is built up in between the two electrodes, behaving like a capacitor. The electron phase shift can be calculated for both regions, according to eq 1, by integrating the resolved spatial-dependent potential along its beam path, and the difference between region (i) and (ii) corresponds to the processed EH phase shifts shown in Figure 2a. According to the theorem of linear summability for electrostatic fields, this procedure equals to calculating the potential difference of the sample area and the reference area before the doing phase integration, which results in the equivalent potential difference model shown in Figure 3b. In the new model, both electrodes are at ground potential, and the effective potentials on the CNT can be derived accordingly. This algorithm requires much fewer finite
element units and gives rise to greater efficiency for calculation without loss of accuracy (details in Supporting Information).

From the in situ I–V measurement, the total resistance across the electrodes shown in Figure 2 is on the order of GΩ, suggesting that either one or both of the two contacts are defective. To uncover the hidden mystery out of the possible three scenarios, three potential difference models are accordingly proposed in Figure 3d in which the top electrode is biased and the bottom one is grounded, just for the consistency with the experiments. After running through all three model calculations (details in Supporting Information), it is found that only the first model, which has one bad contact at the ground electrode, fits the experimental phase-shift data. Figure 4a–d shows the comparison between experimental and simulated phase-shift images acquired under applied bias of +1 V and −1 V, respectively. With the same color coding, the two pairs of phase images display a close agreement in their shapes. Line scans along the arrows across the middle of the corresponding experimental and simulated phase maps are plotted in Figure 4e,f, also verifying their quantitative consistency. It is now apparent that the proposed model for this single-CNT device has been validated, in which the CNT has +1 V constant potential, continuous with the biasing electrode, and an abrupt 1 V potential drop occurs at the ground contact. Using this technique, we are thus capable to locate the break in a nonconducting circuit that does not show any apparent damage, even when the origin of the connection failure is still not clear using high-resolution TEM or other analytical techniques. It is also noted that the asymmetric phase contours of the equiphase contours has distinct correlation with the contact resistances (or potential drops) at the two connection points, as follows: the “fat belly” of the oval tends to be close to the bad (HiR) contact, whereas the narrow end points to the good (LoR) contact. This in fact makes a more important point that EH can be routinely utilized as a generic method to (semi)quantitatively detect contact resistivity within a nanoscale device by a quick capture of electron holograms, which opens up a new approach for localized, uncontacted, and nondestructive diagnosis of nanosystems.

We now turn our attention to another situation to check whether or not EH can distinguish any property change of an electrical contact. Another single-CNT device was fabricated following the same procedure, as displayed in Figure 5a. The bias was applied through the electrode on the top and the other on the bottom was grounded. First, the total resistance of the device was measured as ∼420 Ω at bias of −0.5 V. Then, a voltage of −1 V was applied for a short period of time, until the Pd electrode on the bias side melted due to Joule heating, as shown in Figure 5b. Corresponding electron holograms were recorded before and after the damage, as shown in Figure 5c,d, respectively. According to the asymmetric feature of phase contours in Figure 5d, we can blindly identify a large potential drop at the top electrode, which in fact agrees with the actual connection failure caused by the physically broken Pd electrode. The phase map in Figure 5c also displays a similar asymmetric oval feather, although relatively faint, it indeed indicates a more resistive contact for the top electrode than the one on the other side, predicting the eventual failure at that contact. Further quantification of these two phase-shift images demonstrates that the contact at the biasing electrode exhibits a ∼0.18 V drop out of the total applied 0.5 V bias. It thus gives a reasonable explanation of why overheating occurs at this specific position. In addition, Figure 5c also shows that ∼0.32 V potential drop (∼64% of the total bias) occurs along the length of the nanotube itself, providing an experimental evidence for diffusive conduction of the MWNTs used in this study. This control experiment, as a good example, has justified the validity,
accuracy, and feasibility of the EH technique for its practical utilization in device development and diagnosis. We furthermore add that unlike other TEM imaging methods EH is a relatively noninvasive technique, capable of imaging materials and devices in the natural electronic ground states, as evidenced by studies of low-temperature superconducting metals in an unperturbed superconducting state. The reason for this is due to the electron coherence, which necessarily requires a fairly defocused and low-dose electron beam, giving good plane-wave characteristics, similar to a distant (low-intensity) point-source. This should allow electron holography studies to be extended even to nanomaterials with exotic or fragile electronic ground states.

In conclusion, we have developed a new method to examine the electric potential distribution of a prototype single-CNT device consisting of a suspended CNT with source and drain electrodes, using off-axis EH and finite element calculations. It is demonstrated that two differently resistive contacts gives rise to asymmetric features of the phase shift, and thus can be distinctly identified and quantified. Using this method, we can quickly debug a localized connection failure and detect latent misbehavior on the real device level with a spatial resolution of 10 nm or less. Moreover, the method makes use of a direct and real-space measurement of the electrostatic potential without reliance upon a metallic physical probe that may be susceptible to asymmetric features of the phase shift, and thus can be demonstrated to two diode-like contacts giving rise to asymmetric features of the phase shift, and thus can be distinctly identified and quantified. Using this method, we can quickly debug a localized connection failure and detect latent misbehavior on the real device level with a spatial resolution of 10 nm or less. Moreover, the method makes use of a direct and real-space measurement of the electrostatic potential without reliance upon a metallic physical probe that may be susceptible to variations in the local work function. This approach can be broadly applicable to understanding and diagnosing a variety of nanodevices based on many other nanostructures, such as silicon nanowires and graphene.

**ASSOCIATED CONTENT**

Supporting Information
Detailed description and data analysis of finite element modeling performed by COMSOL. This material is available free of charge via the Internet at http://pubs.acs.org.

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**REFERENCES**


