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Scanning thermal microscopy of carbon nanotubes using batch-fabricated probes

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We have designed and batch-fabricated thin-film thermocouple cantilever probes for scanning thermal microscopy (SThM). Here, we report the use of these probes for imaging the phonon temperature distribution of electrically heated carbon-nanotube (CN) circuits. The SThM images reveal possible heat dissipation mechanisms in CN circuits. The experiments also demonstrate that heat flow through the tip-sample nanoscale junction under ambient conditions is dominated by conduction through a liquid film bridging the two surfaces. With the spatial resolution limited by tip radius to about 50 nm, SThM now offers the promising prospects of studying electron-phonon interactions and phonon transport in low dimensional nanostructures. © *2000 American Institute of Physics.* [S0003-6951(00)03652-4]

In recent years, a number of low dimensional materials with length scale smaller than 100 nm have been developed. Carbon nanotubes,[1] silicon nanowires,[2] and semiconductor/metal nanocrystals[3] are examples of such synthesized nanostructures. There is a great interest to experimentally investigate electron and phonon transport and heat dissipation phenomena in these materials. Such transport and dissipation phenomena are also important in ultra large scale integration (ULSI) devices, whose minimum feature sizes are scaling down to sub-100 nm. Traditional measurement techniques, however, cannot resolve thermal features below 100 nm. For example, the spatial resolution of far-field optical thermometry techniques based on infrared[4] and laser reflectance[5] is diffraction limited to be on the order of wavelength, which is much larger than the length scale of sub-100 nm nanostructures currently of interest.

Scanning thermal microscopy (SThM) is capable of thermally investigating nanostructures and ULSI devices with spatial resolution in the sub-100 nm regime.[6] The SThM maps surface temperature distribution by raster scanning a sharp temperature-sensing tip across the surface. The tip is mounted on a microcantilever beam such that a constant tip-sample contact force is maintained by the force feedback of an atomic force microscope (AFM). Tip-sample heat transfer changes the tip temperature, which is measured and used to determine the sample temperature.

The key element of SThM is the thermal probe. Figure 1 shows the schematic diagram of a SThM probe, which contains a thermocouple junction at the tip end. The tip-sample heat transfer mechanisms include solid–solid conduction through the contact, liquid conduction through a liquid film bridging the tip and sample, and air conduction. The thermal design of the cantilever probes is extremely important for SThM performance. The thermal resistance network in Fig. 1 suggests that for a given ambient temperature, T_a, and sample temperature T_s, the tip temperature T_t, can be written as $T_t = T_s + (T_a - T_s)/(1+\phi)$, and $\phi = R_c/R_{\mathrm{ts}}$ where R_c is the total thermal resistance of the cantilever and the tip, and R_{ts} is the tip-sample thermal resistance. Hence, changes in sample temperature can be related to changes in the tip temperature as $\Delta T_t/\Delta T_s = \phi/(1+\phi)$. This relation suggests that the accuracy and sensitivity of sample temperature measurement by the tip depend on ϕ, which must be large for better SThM performance. The spatial resolution, Δx, of SThM measurements can be expressed as $\Delta x = \Delta T_n/(dT_t/dx)$ where ΔT_n is the noise in the temperature measurement and dT_t/dx is the measured temperature gradient. Because the tip and sample temperatures are related through ϕ, the spatial resolution can be expressed as

$$\Delta x = \frac{\Delta T_n}{(dT_s/dx)} \left(\frac{1+\phi}{\phi} \right). \tag{1}$$

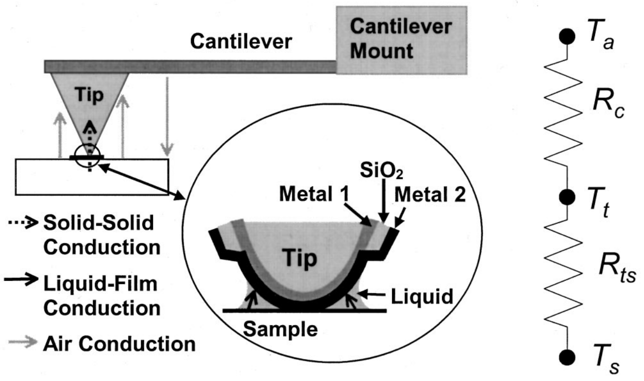

FIG. 1. Schematic diagram of a cantilever probe used for scanning thermal microscopy. The heat transfer mechanisms between the tip, the cantilever and the sample are also indicated. T_a, T_t, and T_s are the temperatures of the ambient, the tip, and the sample, respectively. R_c and R_{ts} are the thermal resistances of the cantilever and the tip-sample junction, respectively.

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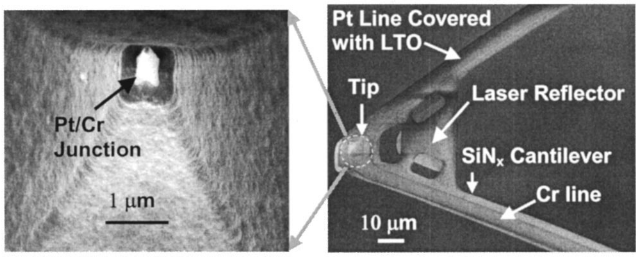


FIG. 2. Scanning electron micrographs of a batch-fabricated probe. Left: a close-up view of the Pt–Cr junction at the tip end; Right: an overview of the probe.

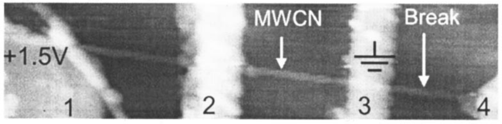
Equation (1) clearly suggests that small values of ϕ lead to poor spatial resolution of SThM.

Previous experiments[7] have reported that $R_{\mathrm{ts}} \approx 10^5$ K/W. Therefore, the thermal design of the cantilever must require $R_c > 10^5$ K/W. In the past, SThM probes were made of a high thermal conductivity material such as metal[8] or silicon,[9,10] with no attention paid to thermal design. This often led to probes with $R_c \ll R_{\mathrm{ts}}$ that led to inaccuracies, loss of resolution, and artifacts.[7] In addition, they were usually fabricated individually,[7,8] making the process very time consuming and irreproducible. Recently, several groups have attempted to batch fabricate probes for scanning thermal microscopy.[9–11] Two groups fabricated thermal probes using only optical lithography and wafer-stage processing steps.[9,10] However, the probes were made of silicon, leading to the aforementioned inaccuracies and artifacts. Silicon nitride thermal probes were also fabricated with a thermocouple junction defined at the tip by electron beam lithography.[11] The low throughput of electron beam lithography prohibits the process for being used for large-volume fabrication.

To address these issues, we have thermally designed and fully batch-fabricated cantilever probes for SThM.[12] Based on heat transfer modeling, we chose silicon dioxide and silicon nitride as the tip and cantilever materials, respectively, so as to increase R_c as much as possible. It was shown by the modeling results that compared to silicon probes with similar geometry parameters, our current design could largely improve the thermal isolation of the sensor from ambient. In addition, platinum and chrome were chosen for the thermocouple materials for their high thermopower difference and low thermal conductivity. Finally, we optimized the geometrical parameters of the probe for increasing thermal resistance of the cantilever and the tip.

The thermal probes were fully batch-fabricated using wafer-stage process steps, with more than 300 probes fabricated on one single wafer.[12] Figure 2 shows two micrographs of a finished cantilever probe with the tip containing a Pt–Cr thermocouple junction, with the Pt and Cr lines patterned along each cantilever arm. The tip region containing the overlap of Pt and Cr thin films was 0.5 μm tall and had a tip radius in the range 20–50 nm. The height of this Pt–Cr junction region could be controlled in the fabrication process to be in the range of 0.1–0.5 μm, with the tip to be 8 μm high. Because the thermal resistance of these probes was very high ($\approx 10^6$ K/W), a low-power laser beam (≈ 1 mW) directed at the tip would increase its tip temperature by up to 80 K. Hence, to optically measure the cantilever deflections for atomic force microscopy, a thermally isolated laser reflector was fabricated.

(a) Topography



(b) Thermal

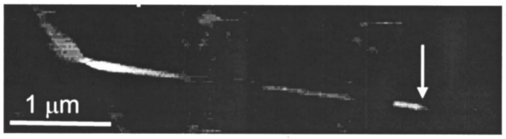


FIG. 3. (a) Topographic and (b) thermal images of a multiwall carbon nanotube circuit under a dc current of 27 μA for an applied voltage of 1.5 V between contacts 1 and 3. Contacts 2 and 4 were floating.

The batch-fabricated thermal probes have been used for quantitative temperature measurement of very large scale integrated via structures[13] and for studying dissipation in multiwall (MW) and single-wall carbon nanotube (CN) circuits.[14] Figure 3(a) shows the AFM topography of one MWCN circuit that was imaged using the SThM probe. The sample contained a 14 nm diameter MWCN and four 30-nm-thick gold contacts on an oxidized silicon wafer. Resistance measurement found that the tube was broken between contacts 3 and 4. The break could barely be identified in a high-resolution AFM image. Figure 3(b) shows the thermal image of the sample obtained for a dc current of 27 μA flowing in the segment between contacts 1 and 3. The temperature rise in the nanotube between contacts 1 and 3 can be clearly observed. To verify that the image was not due to topography-induced artifact, the thermal image was taken at different applied voltages and the thermal signals were found to increase with the voltage. Another possible artifact in the thermal image could be caused by current flow from the sample into the probe due to the difference in electrostatic potentials of the tip and sample. To eliminate this possibility, we measured the tip-sample contact electric resistance when the tip was on top of the CN and the contacts, and found the resistance to be larger than the 1 GΩ measurement range of our ohmmeter for the low contact force used in thermal imaging. In addition, to ensure that no current would flow into the probe even if electric contact established at the tip-sample junction, we connected the thermal probe output to a voltage amplifier with floating ground. We finally confirmed the absence of electrostatic potential-induced artifacts in the thermal images by raising the electrostatic potential of the entire CN circuit without passing current through it. As we did so, no change in the thermal probe output was detected. We, therefore, concluded that the thermal images were indeed due to phonon coupling instead of electron coupling at the tip-sample junction. Furthermore, what the probe measured was the phonon temperature of the sample, which was not necessarily at equilibrium with the electron temperature. Finally, it is important to note that during scanning the probes did not change the currents in the CNs, indicating that the probes did not perturb electron transport in the samples.

It is interesting to note that although no current flowed in the segment between contacts 3 and 4, the temperature of the left part of this segment was higher than that of the segment

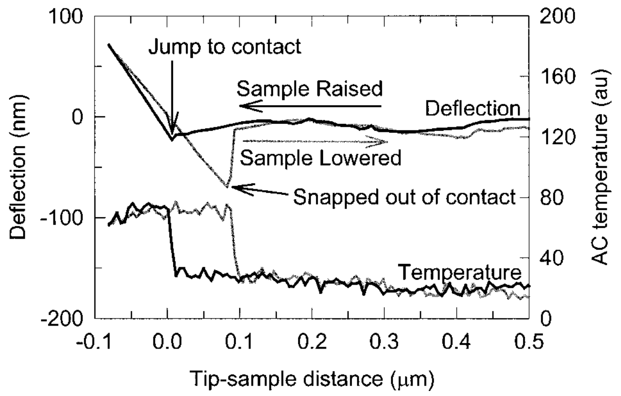


FIG. 4. Cantilever deflection (top) and ac temperature of the thermocouple junction (bottom) as a function of tip-sample distance.

between contacts 2 and 3, where current flowed. Note that switching the polarity of the applied voltage did not change the observed temperature distribution along the tube, indicating that the observed temperature asymmetry was not due to variation in electrostatic potentials. Two mechanisms are suggested by the fact that the CN section between contacts 3 and 4 did not involve any electron transport and yet appeared hotter than the segment involving electron flow. First, more heat might be dissipated at the contacts than in the bulk of the CN. Second, phonon transport is quite efficient in the MWCN from the heated contact to the segment involving no current flow. The temperature between contacts 3 and 4 dropped rapidly at a point indicated by the arrow. We suspect that this was due to phonon scattering by the break, which also blocked electron transport.

This study raises several questions, namely: Does the dissipation really take place at the contacts or in the bulk of the nanotube? How efficient is phonon transport in the nanotube? What are the mean free paths for elastic and inelastic electron and phonon scattering? Some of these questions are currently being addressed experimentally using the SThM and will be presented later.

The full width half maximum of the temperature profile across the CN was on the order of 50 nm, indicating the spatial resolution of the thermal imaging technique. This was approximately equal to the tip diameter. To understand the origin of this superior spatial resolution, the tip-sample heat-transfer mechanisms must be examined. In an earlier work,[7] it was proposed that under ambient condition the tip-sample heat transfer was dominated by conduction through a liquid bridge formed between the tip and sample. However, experimental proof for this hypothesis has remained elusive. To verify the hypothesis, the CN circuit shown in Fig. 3(a) was heated by a 200 Hz ac current and was brought in and out of contact with the thermal probe. The lateral position of the sample was adjusted such that the tip-sample contact point was on the silicon dioxide surface sufficiently close to the heated CN. The ac sample temperature was obtained by measuring the 400 Hz component of the thermal probe output with a lock-in amplifier, and was recorded simultaneously with the cantilever deflection as a function of tip-sample distance, as shown in Fig. 4. Before tip-sample contact occurred, the thermal signal was due to tip-sample heat conduction through the air. In this regime, the thermal signal was low and insensitive to tip-sample distance. When the tip contacted the sample, a liquid bridge formed at the tip-sample junction and pulled the cantilever down, as indicated by ''jump to contact.'' At this point, a large jump in thermal signal can be seen in Fig. 4, indicating significant increase in heat transfer. As the sample was raised further toward the tip, the cantilever was bent up resulting in higher contact force. The thermal signal changed little in this regime, indicating that solid–solid conduction was not a dominant factor in tip-sample heat transfer. As the sample was lowered from the tip, the liquid bridge pulled the tip down until the tip ''snapped out of contact.'' At this point a sudden drop was found in the thermal signal. It is well known that the deflection hysteresis shown in Fig. 4 is due to the presence of a liquid bridge. The coincidence of the thermal hysteresis with that of the deflection clearly indicates that the tip-sample heat transfer was dominated by conduction through the liquid bridge.

In conclusion, we have carefully designed and batch-fabricated probes for scanning thermal microscopy. The probes have been used to obtain thermal images of electrically heated carbon nanotube circuits. Possible heat dissipation mechanisms in the circuits were revealed by the thermal images. Our experimental results also demonstrate that the SThM probes measured the phonon temperature of the sample, and the tip-sample thermal coupling was dominated by heat conduction through a liquid bridge. Consequently, the spatial resolution of SThM was limited by the tip radius and was found to be 50 nm in this study. With this resolution, SThM offers the promising prospects of studying electron-phonon interaction and phonon transport in some low dimensional materials such as carbon nanotubes.

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