Three-dimensional modeling of nanoscale Seebeck measurements by scanning thermoelectric microscopy

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A three-dimensional electrothermal model has been developed to investigate the spatial resolution of the scanning thermoelectric microscopy (SThEM). We found that if the electrical resistivity of the sample changes abruptly, the SThEM will measure a voltage close to the local thermoelectric voltage where electrical resistivity is relatively low, rather than a simple weighted average of the thermoelectric voltage distribution based on the temperature profile. This is due to the presence of internal currents in the sample. The spatial resolution of the Seebeck profiling is limited by the finite value of the phonon mean free path of the sample and the tip size of the microscopy. With a tip size around 1 nm, the scanning thermoelectric microscopy can achieve a spatial resolution of the physical limit defined by the statistical nature of the charge carrier and phonon behavior in a very small region. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2008381]

Thermoelectric devices can remove heat from hot electronic and optoelectronic chips or convert it to electricity without a moving part. The performance of a thermoelectric device is mostly determined by the dimensionless figure of merit ZT. ZT is defined as $ZT=(S^2\sigma/k)T$, where S is the Seebeck coefficient, and σ and k are the electrical and thermal conductivity, respectively. Recently the search for high ZT materials has focused on nanoscale structures due to the enhanced phonon scattering and improved thermoelectric power factor.[1,2] High ZT values of up to 2.4 have been achieved for the thin films and quantum dot superlattices.[3,4]

Since the physical parameters S, σ, and k are modified locally due to the nanoscale features of the materials, the conventional characterization methods for bulk materials may not give accurate results. Scanning thermoelectric microscopy (SThEM) provides a way to measure the Seebeck coefficient directly with nanometer spatial resolution.[5] The setup for Seebeck coefficient profiling is shown in Fig. 1. In an ultrahigh-vacuum environment, the scanning tunneling microscope (STM) worked in the constant current mode. At each point of the measurement, the STM current loop was disconnected and the thermoelectric voltage measurement loop was connected. The STM tip stepped in by the piezocontroller and achieved a nanocontact with the sample. The sample heater caused a temperature difference of 30 K between the sample substrate and the STM tip. A temperature gradient was developed in the sample and localized primarily in the nanocontact region. The thermoelectric voltage V was measured and the local Seebeck coefficient could be calculated by $S=V/\Delta T$.

The measured Seebeck profile of a GaAs p-n junction showed a clear Seebeck coefficient sign change at the transition from the p region to the n region.[5] However, the magnitude of the thermoelectric voltage at the transition region was much lower than the one expected from one dimensional model based on the measured dopant profile. This is attributed to the finite sizes of the tip-sample nanocontact and the temperature gradient around the tip. Because heat diffuses in three dimentions from the sample to the tip, the temperature gradient and Seebeck voltage are distributed in the sample, making up a distributed electrical voltage and resistor network. The measured thermoelectric voltage is the terminal open circuit voltage of this network. In this paper, we present a three-dimensional (3D) electrothermal modeling of the Seebeck profiling of a GaAs p-n junction by scanning thermoelectric microscopy and investigate its spatial resolution for the first time.

The thermoelectric voltage source distribution is determined by the temperature profile and local Seebeck coefficient: $E(x,y,z)=S(x)\Delta T(x,y,z)$. In the bulk materials, the heat conduction is described by continuum equation. However, the phonon scattering mechanism must be taken into

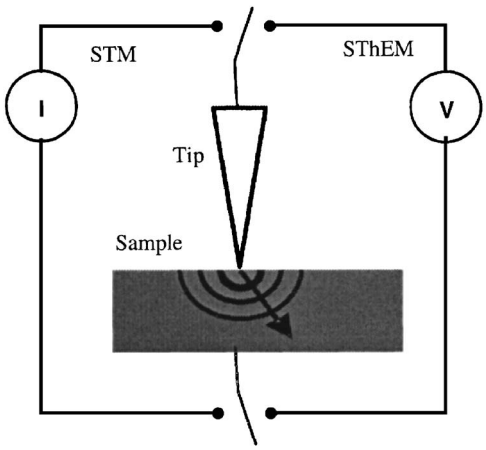

FIG. 1. Schematic of the SThEM setup.

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account for nanostructures because the phonon mean free path is at about the same scale as the structure's dimensions. Thus, we treat the solution of the temperature profile in two regions separately in this model: Around the tip-sample nanocontact and at a distance of a phonon mean free path to the contact, we assume the temperature does not change since there is very little phonon scattering in such a short length. Out of this region, the temperature change is governed by continuum equation of heat conduction. The temperature profile also depends on the shape of the nanocontact. In our model, we assume the contact is a round and flat plate on the surface of the sample. In a phonon mean-free-path distance to the plate, the temperature is assumed to be the same as the plate. Out of this region, the temperature is solved by the finite difference method according to continuum heat conduction. The as-obtained temperature distribution is a first-order approximation of the actual one, which can be obtained using a molecular dynamics or Monte Carlo simulation method.

The Seebeck coefficient can be calculated from the dopant profile. We use the doping density from the experimental data ($N_a=9\times10^{18}$ cm^{-3}, $N_d=1.1\times10^{19}$ cm^{-3}). The electron and hole densities can be solved numerically from the Poisson equation

$$\frac{d^2\phi(x)}{dx^2}=\frac{e}{\varepsilon_0\varepsilon_s}[n(x)-p(x)+N_a(x)-N_d(x)], \tag{1}$$

$$n(x)=n_i\exp\left(\frac{e\phi(x)}{k_BT}\right), \tag{2}$$

$$p(x)=n_i\exp\left(-\frac{e\phi(x)}{k_BT}\right). \tag{3}$$

The Seebeck coefficients from the contribution of electrons and holes are

$$S_e=\frac{k_B}{e}\left[\eta-\frac{\left(r_e+\frac{5}{2}\right)F_{r_e+3/2}(\eta)}{\left(r_e+\frac{3}{2}\right)F_{r_e+1/2}(\eta)}\right],\quad \eta=\frac{E_F-E_c}{k_BT}, \tag{4}$$

and

$$S_h=\frac{k_B}{-e}\left[\eta-\frac{\left(r_v+\frac{5}{2}\right)F_{r_v+3/2}(\eta)}{\left(r_v+\frac{3}{2}\right)F_{r_v+1/2}(\eta)}\right],\quad \eta=\frac{E_v-E_F}{k_BT}, \tag{5}$$

where $F_r(\eta)$ is the Fermi-Dirac integral of order r. For temperatures less than the Debye temperature (344 K for GaAs), the electron and hole scattering parameters $r_e=r_h=1/2$. The total Seebeck coefficient including the contribution of both electrons and holes is

$$S(x)=\frac{n(x)\mu_eS_e(x)+p(x)\mu_hS_h(x)}{n(x)\mu_e+p(x)\mu_h}. \tag{6}$$

The Schottky junction of the tip-sample contact changes the band bending and Seebeck coefficient profile. However, for a nanocontact, the depletion region of the metal-semiconductor junction is also small.[6] If it is smaller than the phonon mean free path, it will only have trivial effects on the thermoelectric voltage measurement. For simplicity, we did not include the effects of tip-sample Schottky junction.

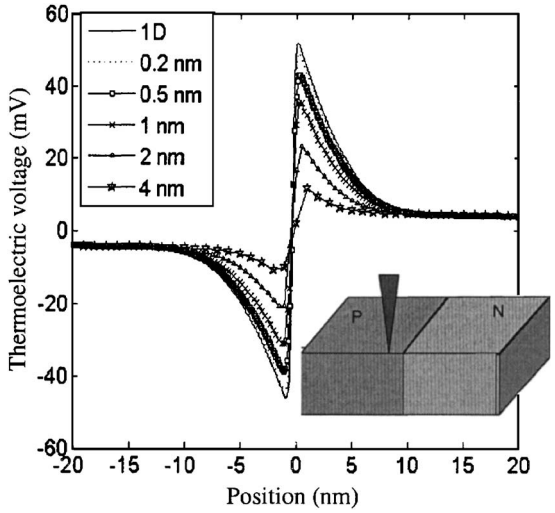

FIG. 2. The comparison of the thermoelectric voltage calculated based on 3D simulations for different phonon mean free paths and the 1D Seebeck profile.

We divided the planar *p-n* junction into $48\times48\times24$ cubic cells of resistor and voltage source network. The resistor network is obtained from the carrier density and the local conductivity of the depletion region and the voltage sources are derived from the 3D temperature distribution and the local Seebeck coefficients. A system of linear equations based on Kirchhoff's laws (conservation of current and voltage) are then written, their solution gives the node potentials. From this, we can calculate the measured thermoelectric voltage at the STM tip.

For a highly doped (10^{19} cm^{-3}) GaAs sample, the phonon mean free path can be smaller than 4 nm. We first neglected the size of the tip contact area and changed the phonon mean free path. The comparison of 3D thermoelectric voltage simulation for different phonon mean free paths and the one-dimensional (1D) Seebeck profile is shown in Fig. 2. The magnitude of the 3D curves is lower than the 1D curve and this explains why the measured Seebeck voltage is lower than the 1D simulation. It can be seen that the magnitude of the 3D thermoelectric voltage profile increases with the decrease of the phonon mean free path and converges to the 1D result. This indicates materials with a smaller phonon mean free path have a higher spatial resolution of Seebeck profiling.

In Fig. 3, we remove the effects of finite phonon mean free path by setting it to zero and change the tip radius alone from 1 to 4 nm. It can be seen that, as expected, a smaller tip size gives a higher spatial resolution of Seebeck profiling. In Fig. 4, we combined the effects of finite phonon mean free path and tip size and set the phonon mean free path to 4 nm for tip radii of 1 and 2 nm. It shows the spatial resolution is further reduced. The measured data are shown as dots, which have similar magnitudes, but the peak location is shifted. The reason for this shift is not very clear. It could be due to nonuniform doping distribution.

It is interesting to note that the measurement of the local thermoelectric voltage is not a simple average of the thermoelectric voltage in the region covered by the tip size and the phonon mean free path. This can be seen from the fact that

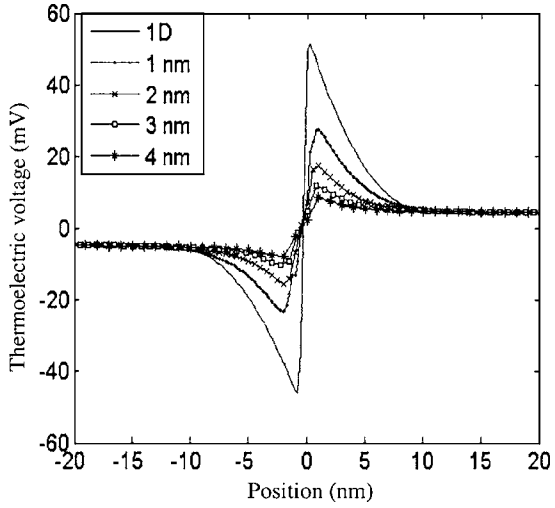

FIG. 3. The comparison of the thermoelectric voltage calculated based on 3D simulations for different tip sizes and the 1D Seebeck profile.

the 3D simulation results are lower than the 1D Seebeck profile almost everywhere in the device. (The 3D curves and the 1D curve should give the same result at a distance far away from the junction). To explain this, we plot the variation of the local thermoelectric voltage and the local resistance of a single grid cell as a function of the distance from *p-n* junction in Fig. 5. When we have a temperature distribution that extends over several nanometers, the contribution of local thermoelectric voltage will be modified due to the nonuniform resistor network. As a simple example in Fig. 5, if we have a voltage source E_1 in series with a resistor R_1 at position 1 and a voltage source E_2 in series with a resistor R_2 at position 2, assuming the tip contact is in the middle of position 1 and 2, the parallel combination of these two sources will produce the following open circuit voltage:

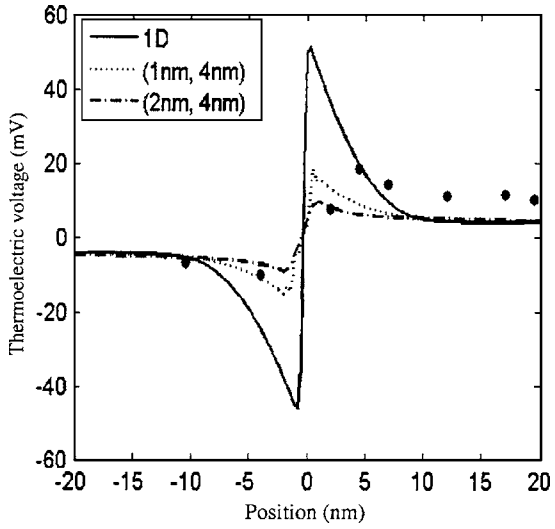

FIG. 4. The 3D thermoelectric voltage simulation for the finite tip size and the phonon mean free path. The dots are experimental data.

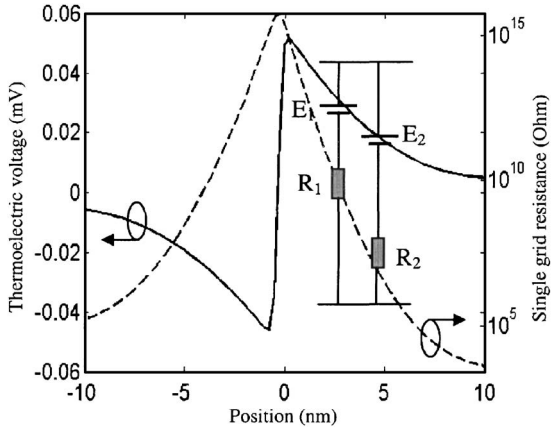

FIG. 5. The 1D thermoelectric voltage and resistance of a single grid cell as a function of distance near the *p-n* junction.

$$V = \frac{R_1 E_2 + R_2 E_1}{R_1 + R_2}. \tag{7}$$

This is due to the internal current flow in the structure. Since the magnitude of the single grid resistance decreases much faster than that of the 1D thermoelectric voltage as we move away from the junction, one should note that the terminal voltage is mainly determined by the lower electrical voltage E_2 which is farther from the junction and in series with the smaller resistance.

In summary, using a 3D model of the thermoelectric voltage measurement by SThEM, we reveal that the 3D effects cannot be simplified as an average of the 1D model of thermoelectric voltage in the adjacent region of the tip-sample contact. The measured value is the terminal voltage of a network of thermoelectric voltages and electrical resisters. The spatial resolution of this nanoscale measurement technique depends on the phonon mean free path, and the size of the tip-sample contact. One should note that the local thermoelectric voltage can only be defined and calculated within a uniform volume larger than the relaxation lengths of the energy/charge carriers and the inter-dopant spacing due to its statistical nature. Thus, this nanoscale Seebeck measurement technique can almost achieve the spatial resolution of the physical limit (the contact size can be made as small as 1 nm and below).

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