# Simultaneous measurements of Seebeck coefficient and thermal conductivity across superlattice

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A method is developed to simultaneously measure the Seebeck coefficient and thermal conductivity in the cross-plane direction of thin films and applied to an *n*-type Si/Ge quantum-dot superlattice. In this method, an Au/Cr pattern serves as both a heater and a thermometer, and a microprobe is prepared between the heater and the thin film to extract the Seebeck voltage. Using a differential measurement between the thin films with different thickness, the temperature and voltage drops across the thin film are determined to deduce its cross-plane thermal conductivity and Seebeck coefficient. At room temperature, the cross-plane Seebeck coefficient and thermal conductivity are 312 $\mu$V/K and 2.92 W/mK, respectively, for the *n*-type Si(75 Å)/Ge(15 Å) quantum-dot superlattice doped to $8.7\times10^{19}$ cm$^{-3}$.  [DOI: 10.1063/1.1458693]

Semiconductor superlattices (SLs) are attracting considerable interests because their low dimensionality may increase the thermoelectric figure of merit.[1–6] Thermoelectric transports in the in-plane (along the film plane) and cross-plane (perpendicular to the film plane) directions of SLs are highly anisotropic and need to be characterized separately. In the cross-plane direction of SLs, electrons and phonons may experience much stronger interface scattering than that along the in-plane direction.[7–9] Furthermore, the thermionic emission process may improve the thermoelectric energy conversion efficiency in the cross-plane direction of SLs.[10–12] To understand thermoelectric transports in the cross-plane direction, one needs to measure, in addition to the thermal conductivity ($k$), the Seebeck coefficient ($S$), and the electrical conductivity ($\sigma$) in the same direction since the device performance depends on the figure-of-merit $Z=S^2\sigma/k$.[13] In the cross-plane direction of SLs, the thermal conductivity has been studied extensively.[7–9,14,15] There are also limited experimental reports for the cross-plane electrical conductivity of SLs.[16] Experiments on the cross-plane Seebeck coefficient in thin films, defined as $S=-\Delta V/\Delta T$, are difficult because it requires the simultaneous determination of the voltage and temperature drops across very thin films ($\sim\mu$m).

In this letter, we report a method for the simultaneous measurement of the cross-plane Seebeck coefficient and thermal conductivity in thin films. This method has been applied to an *n*-type Si/Ge quantum-dot SL.

The principle to measure simultaneously the cross-plane Seebeck coefficient and thermal conductivity of thin films can be explained in Fig. 1(a). The SL layer is fabricated into a mesa structure. The top metal pattern serves as both the heater and the thermometer, similar to the conventional 3$\omega$ method.[17,18] A microprobe between the metal pattern and the SL layer, together with a metal contact pad far away from the heater, is used to extract the cross-plane Seebeck voltage. Since the Seebeck coefficient of the Au lead is much smaller compared to the semiconductor SL and a differential method is used, the contribution of the Au lead to the measured Seebeck coefficient is neglected in our experiment. A reference sample, not shown here, has a structure exactly same as the SL sample except the thinner SL film. A differential measurement between the SL and reference samples is used to eliminate the effects of the substrate, the insulating layers, and interfaces on the voltage and temperature measurements.

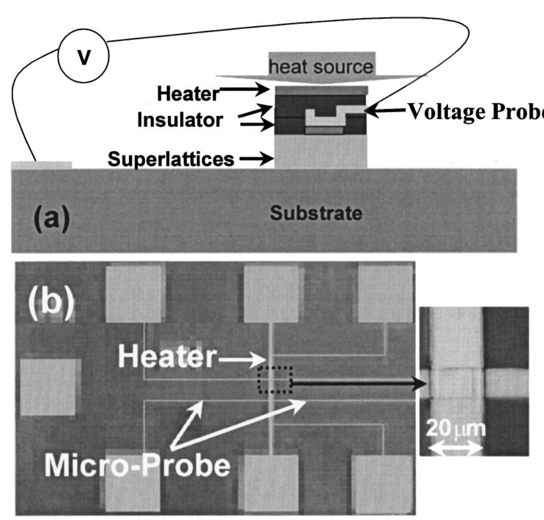


FIG. 1. (a) Schematic illustration of cross section, and (b) photographs of the top surface of the superlattice sample instrumented with heater, temperature sensor, and the voltage probe. The bottom right-hand side picture shows the microprobe underneath the heater.

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If a sinusoidal current at frequency $\omega$ is passed through the top heater, a thermal wave at $2\omega$ will be generated throughout the sample. The temperature oscillation of the heater at $2\omega$ ($T_{2\omega}$) is measured through the small voltage oscillation at $3\omega$ ($V_{3\omega}$) due to the temperature dependent resistance of the heater. The temperature drop across the SL is determined by the measured surface temperatures of the SL and the reference samples. Thus the thermal conductivity of the SL film is given by

$$k_{\rm SL}=P(d_{\rm SL}-d_{\rm reference})/A(T_{2\omega,\rm SL}-T_{2\omega,\rm reference}), \quad (1)$$

where $P$ is the thermal power at $2\omega$, $d$ is the film thickness, $A$ is the area of the heater, and subscript SL or reference denotes the values associated with the SL or the reference sample.[8,18] A $2\omega$ Seebeck voltage wave is also generated by the thermal wave in the samples and detected by the voltage probe embedded underneath the heater. The extra periods in the SL sample should introduce a frequency-independent increase relative to the magnitude of the Seebeck voltage in the reference sample. The magnitude of the cross-plane Seebeck coefficient of SL film can be written in the form

$$|S_{\rm SL}|=|(V_{2\omega,\rm SL}-V_{2\omega,\rm reference})/(T_{2\omega,\rm SL}-T_{2\omega,\rm reference})|. \quad (2)$$

In the experiment, it is found that $\Delta V_{2\omega}$ varies linearly with $\Delta T_{2\omega}$ but doesn't go to 0 while $\Delta T_{2\omega}$ reaches 0. So the cross-plane Seebeck coefficient is determined by $S=d(\Delta V_{2\omega})/d(\Delta T_{2\omega})$ instead of $S=\Delta V_{2\omega}/\Delta T_{2\omega}$. The polarity of the Seebeck coefficient can be deduced from the phase information on the voltage and temperature.

The sample growth and fabrication are briefly described as follows.[19] The Si(75Å)/Ge(15Å) quantum-dot SL samples are grown by solid source molecular-beam epitaxy on a (100) oriented $n$-type Si wafer with electrical resistivity of 0.01 Ω cm. The SL and the reference samples consist of 110 bilayers and 11 bilayers, in which Ge quantum-dot layers (15 Å) are separated by 75 Å-thick Si spacer layers. The SL layers in both samples are uniformly doped with Sb to approximately $8.7\times10^{19}$ cm$^{-3}$. The fabrication process starts with 0.4 $\mu$m-thick plasma-enhanced chemical vapor deposition (PECVD) $SiO_2$ deposition on the sample, whose structure is illustrated in Fig. 1. A small window is opened in this $SiO_2$ layer for the electrical contact of the microvoltage probe with the SL. A layer of 0.2 $\mu$m Au/0.01 $\mu$m Cr is deposited to define the microprobe. A 0.3 $\mu$m thick PECVD $Si_xN_y$ is deposited to insulate the microvoltage probe from the top heater. Reactive ion etching is used to etch away the SL and the insulating layers to make mesa structure.

The raw experimental data for the reference and SL samples are shown in Fig. 2. The slope of the $2\omega$ temperature oscillation curves will yield to the thermal conductivity of the substrate.[17,18] As expected, the presence of the SL thin film produces frequency-independent increases in the $2\omega$ temperature oscillation of the heater (denoted by $\Delta T_{2\omega}$) and in the $2\omega$ Seebeck voltage oscillation (denoted by $\Delta V_{2\omega}$).

Figure 3(a) shows the cross-plane thermal conductivity of the SL, as well as the thermal conductivities for the corresponding bulk Si and Ge (calculated by Fourier's law, $k=0.9k_{\rm Si}k_{\rm Ge}/(0.15k_{\rm Si}+0.75k_{\rm Ge})$,[13] amorphous Si,[18] and $Si_{0.7}Ge_{0.3}$ alloy.[20] The $Si_{0.7}Ge_{0.3}$ alloy is chosen here because it has the optimized composition for thermoelectrics in the SiGe alloy system.[20] As seen in Fig. 3(a), the cross-plane thermal conductivity is lower than the thermal conductivity of SiGe alloy. The huge thermal conductivity reduction in the Si/Ge SL is mainly due to the phonon interface scattering[3] and the decreased phonon group velocity.[2,21]

Figure 3(b) shows the Seebeck coefficient in the cross-plane direction of the SL, as well as the Seebeck coefficients for bulk Si (Ref. 22) and $Si_{0.7}Ge_{0.3}$ alloy[20] with a doping concentration similar to the Si/Ge SL. The temperature dependent behavior of the cross-plane Seebeck coefficient in SL is typical for the doped semiconductor in this temperature range, i.e., a gradual rise in Seebeck coefficient with increasing temperature.[22] In contrast to thermal conductivity, the measured Seebeck coefficient in the cross-plane direction of Si/Ge SL shows only a small change compared to the bulk

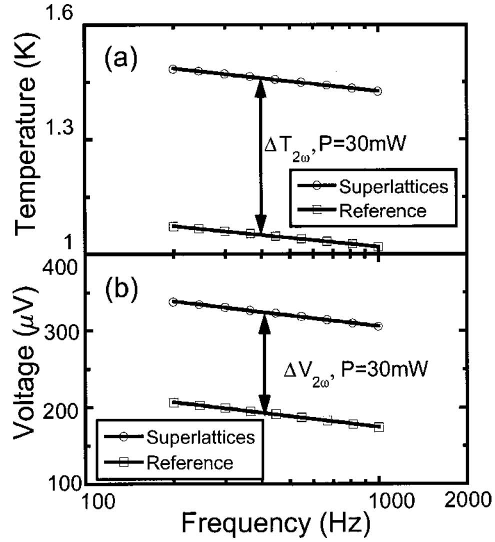


FIG. 2. Measured amplitudes of (a) the temperature and (b) the Seebeck voltage oscillations across the SL sample (circles) and the reference sample (squares) as a function of frequency at 320 K with a heat dissipation of 30 mW. $\Delta T_{2w}$ and $\Delta V_{2w}$ denote the $2\omega$ temperature and voltage drops across the Si/Ge SL film, respectively.

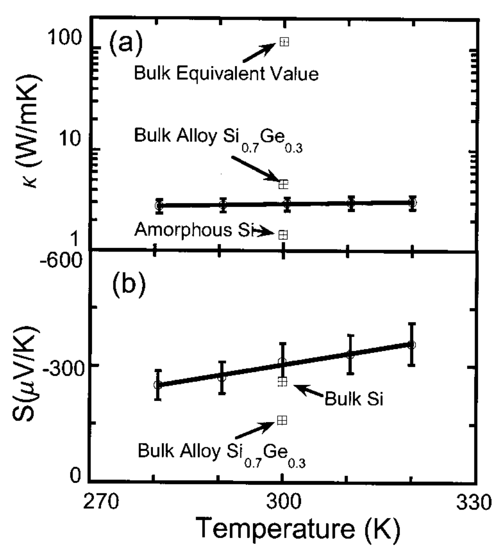


FIG. 3. (a) Thermal conductivity $k$ and (b) Seebeck coefficient $S$ of the $n$-type Si/Ge quantum-dot SL film as a function of temperature. For comparison, the data for amorphous Si, bulk Si and Ge, and bulk alloy $Si_{0.7}Ge_{0.3}$ are also plotted.

values. The possible reason is that the Seebeck coefficient is relatively insensitive to the modification of the electron energy spectrum and the interface scattering, based on the Seebeck coefficient expression in Refs. 1 and 23. Compared to the bulk alloy $Si_{0.7}Ge_{0.3}$, the cross-plane Seebeck coefficient of Si/Ge SL is slightly higher, but the cross-plane thermal conductivity of Si/Ge SL is lower. This suggests that the SL structure is a good candidate for the future high *ZT* thermoelectric materials. In our experiment, the relative uncertainties of Seebeck coefficient and thermal conductivity are about 17%. The uncertainty mainly comes from the temperature rise measurement, calculated from the formula

$$\frac{\delta S_{\mathrm{SL}}}{S_{\mathrm{SL}}} \approx \frac{\delta|T_{2\omega,\mathrm{SL}} - T_{2\omega,\mathrm{reference}}|}{|T_{2\omega,\mathrm{SL}} - T_{2\omega,\mathrm{reference}}|} = \frac{\delta T_{2\omega}}{T_{2\omega}}\left(\frac{T_{2\omega,\mathrm{SL}}}{|T_{2\omega,\mathrm{SL}} - T_{2\omega,\mathrm{reference}}|} + \frac{T_{2\omega,\mathrm{reference}}}{|T_{2\omega,\mathrm{SL}} - T_{2\omega,\mathrm{reference}}|}\right). \tag{3}$$

To cross check this method, the thermal conductivity and Seebeck coefficient of the Si substrate are also determined. They are 131 W/mK and 0.85 mV/K, respectively. These data are in well agreement with the values in literature.[13,22,24]

In summary, a method for the simultaneous measurement of the cross-plane Seebeck coefficient and thermal conductivity in thin films has been developed based on the microfabrication techniques. The Seebeck coefficient and thermal conductivity in the cross-plane direction of an *n*-type Si(75 Å)/Ge(15 Å) quantum-dot SL have been measured. In contrast to the large thermal conductivity reduction, the Seebeck coefficient shows only a small increment in the cross-plane direction of the Si/Ge SL, compared to the corresponding bulk values.

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