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Measurements of anisotropic thermoelectric properties in superlattices

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Thermoelectric properties, i.e., thermal conductivity, electrical conductivity, and the Seebeck coefficient, have been measured in the directions parallel (in-plane) and perpendicular to the interface of an *n*-type Si(80 Å)/Ge(20 Å) superlattice. A two-wire 3ω method is employed to measure the in-plane and cross-plane thermal conductivities. The cross-plane Seebeck coefficient is deduced by using a differential measurement between the superlattice and reference samples and the cross-plane electrical conductivity is determined through a modified transmission-line method. The in-plane thermal conductivity of the Si/Ge superlattice is 5–6 times higher than the cross-plane one, and the electrical conductivity shows a similar anisotropy. The anisotropy of the Seebeck coefficients is smaller in comparison to electrical and thermal conductivities in the temperature range from 150 to 300 K. However, the cross-plane Seebeck coefficient rises faster with increasing temperature than that of the in-plane direction. © *2002 American Institute of Physics.*
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Semiconductor superlattices (SLs) are attracting much attention because their low dimensionality may increase the thermoelectric figure of merit.[1–7] Theories have been developed on both electron and phonon transports in SLs.[1–4,8–11] Experimental characterization of thermoelectric properties has proven to be very challenging because these properties are highly anisotropic and need to be characterized for either or both directions parallel (in-plane direction) and perpendicular (cross-plane direction) to the SL interfaces. Thermoelectric properties include thermal conductivity (k), electrical conductivity (σ), and the Seebeck coefficient (S) that determine the figure of merit $Z=S^2\sigma/\mathrm{k}$.[12] The cross-plane thermal conductivity of SLs has been extensively studied,[13–17] however, there are less experimental reports on the in-plane thermal conductivity of SLs because of the heat leakage through the substrate in the measurements.[7,18–20] Measuring the electrical conductivity and Seebeck coefficient in the cross-plane direction is particularly difficult because of the small film thickness ($\sim\mu$m).[6,21] Most previous experimental studies on thermoelectric thin films such as SLs were carried out in separate samples or different directions. It is highly desirable to characterize all thermoelectric properties in both directions on the same sample.

In this letter, we report an experimental study on the thermoelectric properties, i.e., thermal conductivity, electrical conductivity, and the Seebeck coefficient, in both in-plane and cross-plane directions of an *n*-type Si(80 Å)/Ge(20 Å) SL. The anisotropy and the temperature dependence of these properties will be discussed.

The experimental methods used in this letter are described briefly here.[6,20–23] Two samples, named the SL sample and the reference sample, are used in the differential measurement to eliminate the effects of the substrate, the buffer layer, and the interfaces since the reference sample has the same structure as the SL sample except a thinner SL film, as shown in Fig. 1. The SL film is grown on a silicon-on-insulator (SOI) wafer to ensure the electrical insulation between the SL film and the substrate, which is necessary for the SL electrical conductivity measurement. To measure the anisotropic thermal conductivity, a two-wire 3ω method is applied to the SL sample.[20,22] Two heaters with different wire widths are deposited on the top of the sample. The one having a wire width much larger than the film thickness is used to determine the temperature drop due to cross-plane heat conduction. The one with a width smaller or comparable to the film thickness will measure the temperature drop due to both in-plane and cross-plane heat conduction. The in-plane and cross-plane thermal conductivities can be extracted from the temperature rise data for these two wires.

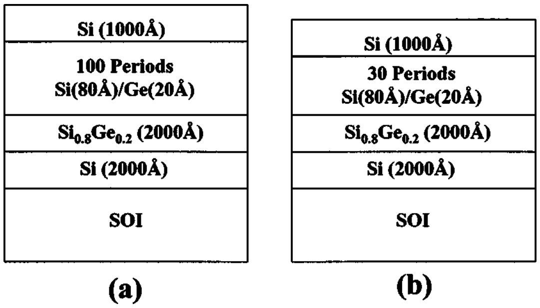

FIG. 1. Configurations of the SL sample (a) and the reference sample (b).

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To determine the cross-plane Seebeck coefficient of the SL film, the temperature and voltage drops across the film need to be measured at the same time.[21] A Au/Cr pattern, deposited on the top of SL film, 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 SL sample and the reference sample, the temperature and voltage drops across the thin film are measured to deduce its cross-plane Seebeck coefficient.

The cross-plane electrical conductivity of the SL film is obtained by a modified transmission-line method (TLM) that is a classical approach for the semiconductor–metal contact resistance measurement.[6,23] The modification taken here is to etch down the SL film except the area underneath the metal contact line to make a mesa structure. In this SL mesa structure, the measured total contact resistance includes the Si–metal contact resistance and the cross-plane SL resistance. Mesa structures with different heights are used to eliminate the effect of the SL–metal contact and to obtain the cross-plane SL resistivity. One critical condition for the cross-plane resistivity measurement is that the contact resistance should not overwhelm the cross-plane SL resistance. In our experiment, the contact resistance is large relative to the SL resistance near room temperature and below, which limited the cross-plane electrical conductivity measurements to the elevated temperatures.

The conventional four-point-probe method is employed to measure the in-plane electrical conductivity.[22,23] The in-plane Seebeck coefficient is also measured in the same structure while a temperature gradient is applied along the sample.

The samples were grown by solid-source molecular beam epitaxy on (100)-oriented SOI wafers. To obtain the symmetrically strained Si/Ge SL, a relaxed SiGe buffer is needed. In this approach, we used low-temperature-Si assisted SiGe layer growth.[24] A 2000-Å-thick Si buffer layer was grown at 400 °C, followed by a 2000-Å-thick $Si_{0.8}Ge_{0.2}$ alloy layer grown at 550 °C. The SL film contains a 100-period Si(80 Å)/Ge(20 Å) layer, uniformly doped with Sb to $\sim 10^{16}$ cm^{-3}. This low doping is chosen based on the minimum contact resistance we can realize and the estimated cross-plane electrical conductivity. For the purpose of making Ohmic contact, a 1000-Å-thick Si layer with a doping concentration of about 10^{20} cm^{-3} is grown on the top of the SL layer. A reference sample, which has exactly the same structure as the SL sample except a thinner SL layer (30 periods), is prepared to eliminate the effects of the substrate, the buffer layer, and the interfaces in the differential measurements. The structures of the SL and the reference sample are shown in Fig. 1. In the thermal conductivity measurement, the Si cap layer is etched away by a reactive ion etcher to avoid heat spreading.

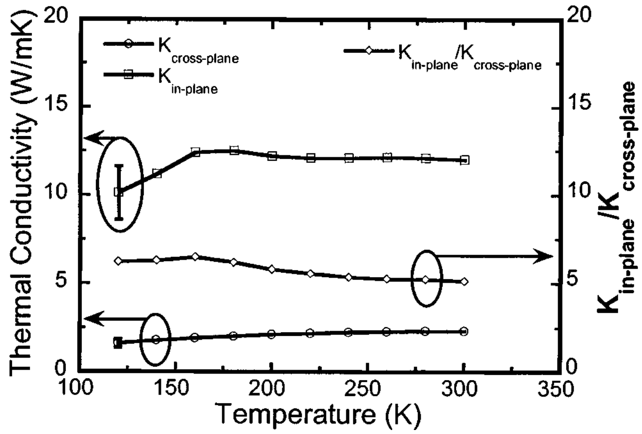

FIG. 2. In-plane and cross-plane thermal conductivities of the Si(80 Å)/Ge(20 Å) SL as a function of temperature.

The in-plane and cross-plane thermal conductivities of the Si(80 Å)/Ge(20 Å) SL are shown in Fig. 2. The anisotropy and the temperature dependence of thermal conductivity can be used to infer the effects of various phonon scattering mechanisms, which depend on temperature in different ways. It is well known that the bulk Si and Ge thermal conductivities go as $T^{-\alpha}$ (where α has a value of about 1.65 for Si and 1.25 for Ge) in the high-temperature range due to the predominating umklapp scattering and normal three-phonon scattering.[25,26] The thermal conductivities of the Si/Ge SL, however, do not show a similar temperature dependence. In the cross-plane direction, the temperature dependence follows the specific heat behavior. This implies that temperature-dependent scattering mechanisms, such as umklapp scattering, do not contribute too much to heat conduction, and interface scattering, which is insensitive to temperature should dominate the transport. In the in-plane direction, the SL thermal conductivity decreases slowly with increasing temperature, and a peak appears at a temperature much higher than that in bulk materials. These suggest that for the in-plane heat conduction, the interface still plays an important role while the normal three-phonon scattering and umklapp scattering gradually set in with increasing temperature. The anisotropy of the thermal conductivity is mainly attributed to the interface scattering[3] and SL phonon velocity.[2,9,27] The ratio of the in-plane to the cross-plane thermal conductivities decrease with increasing temperature because the umklapp scattering and normal three-phonon scattering become more important in the high-temperature range.

The in-plane electrical conductivities of the Si(80 Å)/Ge(20 Å) SL are shown in Fig. 3. Due to the contact resistance issue, the cross-plane electrical conductivity is measured by the modified TLM only at 480 K. At this temperature, the cross-plane and in-plane conductivities are 117 and 580 $\Omega^{-1}\,m^{-1}$, respectively. The in-plane electrical conductivity at $T \leqslant 300$ K is measured by the conventional four-point-probe method. The temperature dependence of the in-plane electrical conductivity in the SL is typical for the lightly doped semiconductor in this temperature range, i.e., the electrical conductivity declines with decreasing temperature.[28] The ratio of the in-plane electrical conductivity to the cross-plane electrical conductivity at $T = 480$ K is about 5, similar to the thermal conductivity. It is worth noting that the electrical conductivity anisotropy of the heavily doped Bi_2Te_3/Sb_2Te_3 SL is much lower according to a recent report.[6] The possible reason for the anisotropy difference between the Si/Ge SL and the Bi_2Te_3/Sb_2Te_3 SL is that the conduction-band offset in Bi_2Te_3/Sb_2Te_3 SL is smaller than that in the Si/Ge SL. Also, the high doping concentration may benefit more the cross-plane electron transport be-

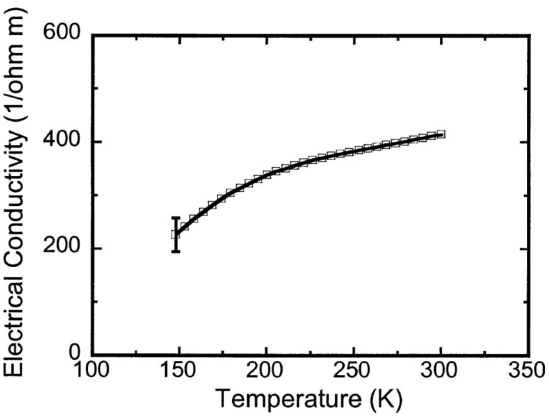

FIG. 3. In-plane electrical conductivities of the Si(80 Å)/Ge(20 Å) SL as a function of temperature. At 480 K, the measured cross-plane and in-plane electrical conductivities are 117 and 580 $\Omega^{-1}\,\mathrm{m}^{-1}$, respectively, leading to an anisotropy (in plane divided by the cross plane) of 4.96.

cause the energy difference between the Fermi energy and the conduction band of the barrier layer is smaller in the *n*-type SL with higher doping concentration.

The in-plane and cross-plane Seebeck coefficients of the Si/Ge SL are shown in Fig. 4. In contrast to thermal conductivity and electrical conductivity, the measured Seebeck coefficient does not show a high anisotropy. Seebeck coefficients in both directions gradually rise with increasing temperature, but their slopes with respect to temperature are different. The cross-plane Seebeck coefficient increases faster with temperature and tends to be larger than the in-plane Seebeck coefficient in the high-temperature range. It has been theoretically reported that the Seebeck coefficient in two-dimensional structures, such as semiconductor SLs, is isotropic based on the assumption of isotropic scattering.[9] The discrepancy between the theoretic prediction and the measured data indicates that electron scattering in the SL should be high anisotropic, which may be accounted for by anisotropic interface scattering. Since the investigated sample is low doped, the absolute ZT of the SL is low. The cross-plane ZT is about 1.2 times higher than the in-plane ZT at $T=300$ K, assuming $\sigma_{\text{in-plane}}/\sigma_{\text{cross-plane}}=5$. $ZT_{\text{cross-plane}}/ZT_{\text{in-plane}}$ should become larger at the higher temperatures since the cross-plane Seebeck coefficient increases faster with temperature.

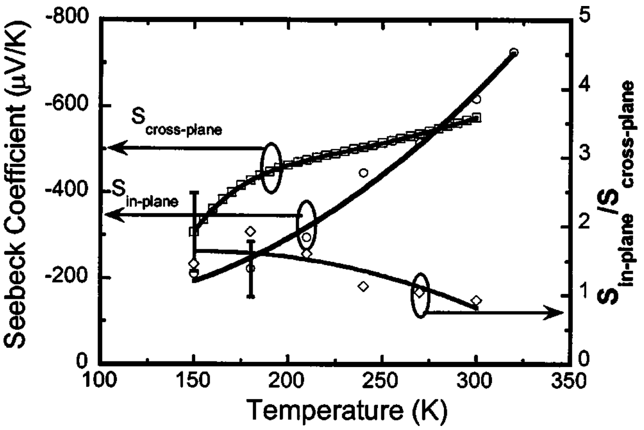

FIG. 4. In-plane and cross-plane Seebeck coefficients of the Si(80 Å)/Ge(20 Å) SL as a function of temperature.

In summary, the cross-plane and the in-plane thermoelectric properties of an *n*-type Si/Ge SL have been measured and their anisotropy and temperature dependence have been studied. Both thermal and electrical conductivities show a large anisotropy. In contrast, the Seebeck coefficient does not show a large anisotropy in the temperature range from 150 to 300 K, but the cross-plane Seebeck coefficient depends more strongly on temperature than that of the in-plane direction.

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