Study of thermomechanical properties of Si/SiGe superlattices using femtosecond transient thermoreflectance technique

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Using a Femtosecond Transient Thermoreflectance (FTT) technique, we studied the thermomechanical properties of two Si/SiGe superlattices. A theoretical model is presented which agrees well with the experimental results and allows us to determine the cross-plan thermal conductivity of the superlattices at room temperature. We also show that, from the experimental curve, we can extract the thickness of the metallic film, the longitudinal sound velocity and the refractive index of the superlattice using acoustic echoes and Brillouin oscillation data. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2009069]

Several experimental studies have pointed out that the thermal conductivity of semiconductor superlattices (SLs) is lower than that of the bulk material.[1–4] Several theoretical studies have tried to explain these observations, and many models have been developed.[5,6] These structures, which alternate different semiconductor thin layers, have proved to have not only a low thermal conductivity but also a high thermoelectric power, which makes them potential candidates especially in thermoelectric area.

The thermal conductivity of semiconductors comes essentially from the lattice contribution, and it is limited by the rate at which phonons are scattered. There are two different scattering processes that phonons can undergo: an *intrinsic* process arising from the anharmonicity of the interatomic forces, and an *extrinsic* process that is due to the phonons scattering because of various sorts of crystal defects, and crystal surface (impurities, grain boundary, etc.). Peierls[7] pointed out that the anharmonic processes were of two types, Normal processes (N-process) and UMKLAPP processes (U-process). For an N-process, the vector sum of the phonon momenta is unchanged after a collision, whereas for a *U*-process, the total momentum changes by a reciprocal-lattice vector. In a bulk crystal, there is a minimum phonon energy required for a U-process to occur. In an SL the minimum phonon energy is lower because the magnitude of the shortest reciprocal-lattice vectors is smaller.[8]

In the experiments reported here we have applied a FTT technique to study two Si/SiGe SLs, $Si/Si_{0.7}Ge_{0.3}$, and $Si/Si_{0.4}Ge_{0.6}$. Due to the lattice mismatch between silicon and germanium, which is about 4.2%, doping silicon by more than 40% will increase the number of defects and dislocations. As a consequence, the structure will not be stable as it is reported by Douglas.[9] The defects interact with the electrical, optical and thermal properties of the material, typically degrading their performances.[9]

The first sample is a 1 μm *n*-type superlattice [$67 \times (12\text{ nm } Si_{0.7}Ge_{0.3}/3\text{ nm Si})$] doped to 6.8×10^{19} cm^{-3}, and grown on top of 0.15 μm $Si_{0.8}Ge_{0.2}$ buffer layer with silicon on insulator substrate. The second one is a 3 μm *p*-type superlattice [$246\times(4\text{ nm } Si_{0.4}Ge_{0.6}/8\text{ nmSi})$] doped to 5×10^{19} cm^{-3}, and grown on top of the ($1\ \mu\text{m } Si_{0.8}Ge_{0.2}/0.6\ \mu\text{m } Si_{0.9}Ge_{0.1}/Si_{0.845}Ge_{0.15}C_{0.005}$) buffer layer with silicon substrate. In both samples, the buffer layer is used to reduce mechanical stress due to the lattice mismatch of the SL with respect to silicon. In fact this layer acts as a "*virtual substrate*." A 12 nm thick aluminum film is then deposited on the samples surface. Figure 1 shows a schematic diagram of the experiment. A pump light pulse, focused to a small spot on the surface of the metallic film, creates a sudden temperature rise. A small change in the temperature of the film $\Delta T(t)$ produces a proportional change in the optical reflectivity[2] $\Delta R(t)$. As the film cools by conduction into the underlying SL, the change in the reflectivity is measured by means of a time-delayed probe light pulse which is focused onto the film so as to overlap with the pump pulse. In order to determine the cross-plan thermal conductivity $\beta_s^{\perp}$ of the SL at room temperature ($T=300$ K), the change in the reflectivity as a function of delay time is compared to the change calculated from theoretical modeling. The value of $\beta_s^{\perp}$ used in simulation is adjusted so as to give the best fit to experimental data.

The light pulses are produced by a Ti:sapphire laser operating at a repetition rate of 82 MHz, a wavelength of 780 nm, and pulse duration of 100 fs. The pump beam is chopped at a frequency of 700 kHz by an Acousto-Optic

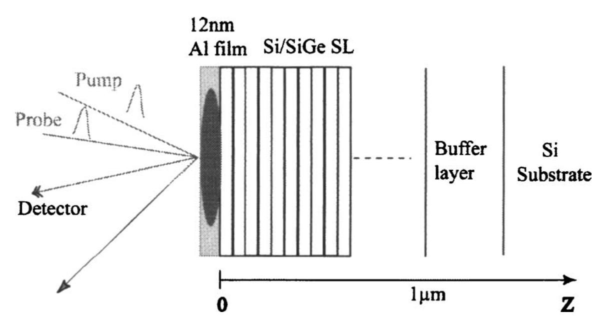

FIG. 1. Schematic diagram of the experiment and sample structure.

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Modulator (AOM), and focused to a spot of about 20 μm. The probe beam is focused to a spot of about 6 μm on the same region of sample illuminated by the pump. The optical path length of the probe pulse is varied relative to the optical path length of the pump pulse by reflecting the beam off a retroreflector mounted on a mechanical translation stage. The energies in each pump and probe pulse applied to the film are typically 1.4 nJ, and 0.2 nJ, respectively. To improve the signal to noise ratio, the output of the detector, which detects the reflected probe beam, is amplified by a lock-in amplifier, which is locked at 700 kHz.

When the pump pulse is absorbed, its energy is at first communicated to electrons near the front surface of Al film which will be excited to higher energy states. These hot electrons quickly diffuse away from the Al surface, but are confined to the Al film by the Schottky barrier at the interface Al/SL. Within several picoseconds, the hot electrons transfer their energy to the lattice by electron-phonon collisions, slightly raising the temperature of the Al film. The temperature distribution inside the Al film becomes uniform by thermal diffusion, and phonons escape across the interface into the SL.[10] The diffusion of the phonons across the interface is relatively slow compared to the thermal transport within the Al film.[2]

The time delay of the experiment was about 1 ns, so the analysis of the cooling of the Al thin film into the Si/SiGe SL is greatly simplified because it is possible to neglect radial flow in the Al film and the SL, since the thermal diffusion length is much smaller than the radius of the focused pump beam. The problem is then one-dimensional in the normal direction of the Al film surface. Furthermore, the SL will act as a semi-infinite medium, and then neither the buffer-layer nor the silicon substrate will influence the cooling rate of Al film. The thickness of the Al film (12 nm), is about twice the optical penetration depth of Aluminum, $\xi = 7$ nm @ $\lambda = 780$ nm, then, all the film will be heated at the same time and the temperature distribution will be uniform inside it. A few percent of light crosses the Al film, and penetrates the SL. This effect is not taken into account in the model.

Heat transfer in the cross-plan direction of the structure is governed by the following equations:

$$C_f d_f \frac{\partial T_f}{\partial t} = \beta_s^{\perp} \left. \frac{\partial T_s}{\partial z} \right]_{z=0}. \tag{1}$$

This describes the energy conservation at the interface Al/SL. C_f and d_f are, respectively, the specific heat per unit volume (in $J/K/m^3$), and thickness of the Al film, $\beta_s^{\perp}$ is the cross-plan thermal conductivity of the SL.

The interface Al/SL acts as a thermal barrier and then, there is a jump of temperature given by

$$T_f - T_s = -\beta_s^{\perp} R_k \left. \frac{\partial T_s}{\partial z} \right]_{z=0}. \tag{2}$$

R_k is the thermal boundary resistance or Kapitza resistance at this interface.

In the SL,

$$\frac{\partial^2 T_s}{\partial z^2} = \frac{1}{\alpha_s^{\perp}} \frac{\partial T_s}{\partial t}, \tag{3}$$

where $\alpha_s^{\perp}$ is the cross-plan thermal diffusivity of the SL.

The initial condition is given by

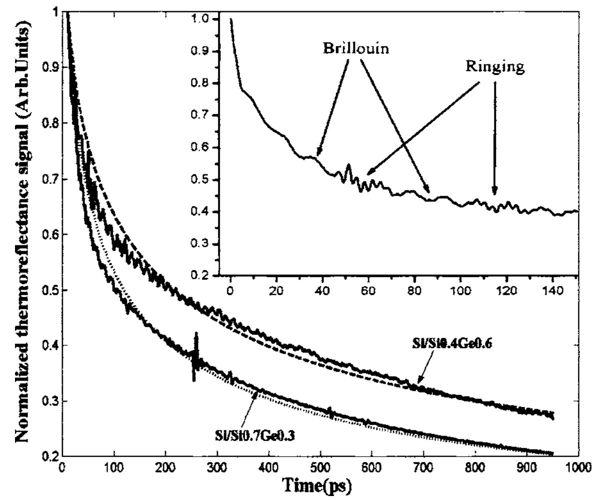

FIG. 2. Comparison between normalized experimental thermoreflectance signal (solid line), and theoretical modeling for the best fit of the two superlattices: $Si/Si_{0.4}Ge_{0.6}$ (dashed line), and $Si/Si_{0.7}Ge_{0.3}$ (dotted line). The inset shows the two types of oscillations obtained on the $Si/Si_{0.4}Ge_{0.6}$ SL: Brillouin oscillation with a frequency $f_{\mathrm{Brillouin}}^{\mathrm{Exp}} \simeq 72$ GHz, and ringing oscillation with a frequency $f_{\mathrm{ringing}}^{\mathrm{Exp}} \simeq 276$ GHz.

$$T_f(t=0) = \frac{(1-R)Q}{C_f d_f A_f}. \tag{4}$$

$R \approx 0.9$ is the reflection coefficient of Al at the wavelength of the experiment, $Q \approx 1.4$ nJ is the pump pulse energy, and $A_f = \pi r^2$ is the illuminated area of the Al film surface, $r \approx 10$ μm is the radius of the focused pump beam. Taking room temperature as temperature reference, an estimated value of the initial temperature variation is $T_f(t=0) \approx 15$ °C.

On the other hand, due to the small thermal diffusion length, heat penetrates only few tens nm into the SL, and so

$$T_s(z=\infty) = 0. \tag{5}$$

Resolution of this system of equations is more simplified in Laplace domain, and we found the following solution:

$$\theta_f = \frac{\tau_{sf} + \tau_R \sqrt{p\tau_{sf}}}{\sqrt{p\tau_{sf}} + p(\tau_{sf} + \tau_R \sqrt{p\tau_{sf}})} T_f(t=0), \tag{6}$$

where p is the Laplace variable, $y = C_s/C_f$, $bi = d_f/\beta_s^{\perp} R_k$, $\tau_{sf} = d_f^2/\alpha_s^{\perp} y^2$, and $\tau_R = \tau_{sf} y/bi$. C_s, is the specific heat per unit volume of the SL. τ_{sf} is a diffusive time which describes the diffusion of heat inside the SL, τ_R is a resistive time which describes the thermal barrier behavior of the interface Al/SL.

Figure 2 shows the thermoreflectance signal of the Al film for two different SLs $Si/Si_{0.7}Ge_{0.3}$ and $Si/Si_{0.4}Ge_{0.6}$. The signals present one thermal decay which describes the diffusion of heat inside the SL, and on this thermal decay are superimposed some spikes, features of acoustic echoes at different interfaces inside the structures. We have studied the sensitivity of the temperature variation on the Al film top surface using numerical Laplace inversion.[11] This study has shown that the Al film cooling is more sensitive to the thermal conductivity of the SL than to the Kapitza resistance at the interface Al/SL. Moreover, the value of this resistance was found to be smaller than 5×10^{-10} m^2 K/W, this value is about one order of magnitude smaller than the values of literature.[12] We can then consider a perfect thermal interface between the Al film and the Si/SiGe SL, in this case $\tau_R \to 0$, and Eq. (6) becomes

$$\theta_f = \frac{\tau_{sf}}{\sqrt{p\tau_{sf}} + p\tau_{sf}} T_f(t=0). \quad (7)$$

A simple analytical solution of the cooling rate of Al film can be obtained, using Laplace inverse transform table[13]

$$T_f(t) = T_f(t=0)\exp\left(\frac{t}{\tau_{sf}}\right)\mathrm{erfc}\left(\sqrt{\frac{t}{\tau_{sf}}}\right). \quad (8)$$

Using the Least Square Method (LSM) to optimize the theoretical curve to fit the experimental data gives the following values: $\beta^{\perp}_{Si/Si_{0.7}Ge_{0.3}} \simeq 8.1$ W m^{-1} K^{-1}, and $\beta^{\perp}_{Si/Si_{0.4}Ge_{0.6}} \simeq 2.8$ W m^{-1} K^{-1}, for the cross-plan thermal conductivity of $Si/Si_{0.7}Ge_{0.3}$, and $Si/Si_{0.4}Ge_{0.6}$, respectively. These values are in good agreement with those found in the literature.[14,15] Using these values, we get the best fit to the experimental data shown in Fig. 2. The small discrepancy between theoretical curves and experimental signals comes from the fact that 18% of light energy crosses the Al film and reaches directly the SL, which is not taken into account in the model discussed above. The thermal conductivity of $Si/Si_{0.4}Ge_{0.6}$ SL is inferior to that of $Si/Si_{0.7}Ge_{0.3}$ SL. This result was expected, since in $Si/Si_{0.4}Ge_{0.6}$ SL, phonons undergo much more scattering processes than in $Si/Si_{0.7}Ge_{0.3}$ due to the presence of various crystal defects and surface defects, caused by the lattice mismatch, and high Germanium concentration, Therefore, it leads to an increase of extrinsic scattering process and then reduces the phonon mean free path and consequently the thermal conductivity.

In Fig. 2, we can see some echoes and some oscillations superimposed to the thermal decay. The knowledge of the arrival time of the first echo in conjunction with the thickness of the $Si/Si_{0.7}Ge_{0.3}$ SL allows us to get the longitudinal sound velocity inside the later. We found a value of $v^{Exp}_{Si/Si_{0.7}Ge_{0.3}} \simeq 7822$ m/s, this value is in good agreement with the estimated value using the harmonic average,[16] $v^{The}_{Si/Si_{0.7}Ge_{0.3}} \simeq 7078$ m/s. The inset shows another experimental curve obtained on $Si/Si_{0.4}Ge_{0.6}$ at the first 200 ps time scale, where we can distinguish two types of oscillations. But what is the origin of these oscillations?

After the impact of the light pulse, a part of the energy (about 18%), crosses the Al film and then reaches directly the underlying SL. An acoustic pulse is then produced, and propagates in the SL. The interaction of the probe beam with this acoustic wave will produce the first type of oscillations, called "*Brillouin oscillations*." As first observed by Thomson *et al.*,[17] the probe light pulses are reflected on the moving acoustic wave front and produce interferences with the first interface reflection, just like in a Fabry-Perot cavity. The oscillation frequency is related to the velocity of the acoustic wave front. In fact light is retrodiffused by phonons, so this retrodiffusion will get both constructive and destructive interferences between successive reflected probe pulses. On the other hand, the quantity of light absorbed in the Al film (about 82%) will heat it suddenly, initiating oscillations inside the structure. The reflection of this acoustic pulse, *called* "*ringing*," at an interface gives the observed oscillations in the thermoreflectance signal. Measurements give the following frequencies of these two types of oscillations: $f^{Exp}_{Brillouin} \simeq 72$ GHz, and $f^{Exp}_{ringing} \simeq 276$ GHz. Then, we can extract both Al film thickness, and the product $n_{Si/Si_{0.4}Ge_{0.6}} v_{Si/Si_{0.4}Ge_{0.6}}$. Indeed, theoretical expression of the ringing frequency is given by $f^{Th}_{ringing} = v_f / 2d_f$.. So, if we assume bulk sound velocity at room temperature for the Al film $v_f \simeq 6400$ m/s, we find $d_f \simeq 11.6$ nm. In the same way, the Brillouin frequency is given by

$$f^{Th}_{Brillouin} = \frac{2n_{Si/Si_{0.4}Ge_{0.6}} v_{Si/Si_{0.4}Ge_{0.6}} \cos(\theta)}{\lambda}, \quad (9)$$

where $n_{Si/Si_{0.4}Ge_{0.6}} v_{Si/Si_{0.4}Ge_{0.6}}$ λ, and θ are, respectively, the index of refraction of the SL at the wavelength of the experiment, the longitudinal sound velocity inside the SL, the wavelength of the experiment, and the incidence angle of the probe beam. In our experiment, the incidence was normal, so from the measurement value, we can get $n_{Si/Si_{0.4}Ge_{0.6}} v_{Si/Si_{0.4}Ge_{0.6}} \simeq 28080$ m/s. Furthermore, if we assume the value of the sound velocity of the SL estimated using the harmonic averge,[16] we found $v^{The}_{Si/Si_{0.4}Ge_{0.6}} \simeq 7284$ m/s, and then, we obtain the value of the index of refraction of the SL at the wavelength of the experiment: $n_{Si/Si_{0.4}Ge_{0.6}} \simeq 3.85$. This value is well in the range of values of the index of refraction of semiconductors at this wavelength.[18]

The cross-plan thermal conductivity of two different Si/SiGe superlattices was identified using a FTT technique. This identification was made possible by fitting the experimental data with a theoretical calculation of the Al film cooling using a LSM. We have shown how the defects inside the structure can considerably reduce the thermal conductivity from alloy limits. We were also able to extract the longitudinal sound velocity of one of the underlying superlattice and the index of refraction of the other at the wavelength of the experiment from the measured Brillouin oscillation frequency. When the film is very thin, the pump laser pulse produces ringing oscillation from which, we could measure the film thickness.

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