I. Sample growth and structural properties

All investigated samples were grown by solid source molecular beam epitaxy (MBE). After RCA cleaning and in situ deoxidation, a 100 nm thick Si buffer was grown, followed by stacks of Ge layers separated by Si spacers (with thickness $t_{Si} = 3, 6, 9$ and 12 nm) deposited at a nominal substrate temperature of 500°C. The Ge layer on the sample surface was used for atomic force microscopy (AFM) characterization. Two sample sets were studied here: set #1 contains 5 and set #2 11 Ge layers (see Fig. 1a). The first island layer was obtained by deposition of 6.4 monolayers (ML) or 5.8 ML of Ge (for set #1 and #2, respectively), leading to the formation of $\{105\}$ faceted “hut clusters” [Mo90] (see scanning tunneling microscopy – STM image in Fig. SM1) with an average height of $1.2 \pm 0.2$ nm and surface densities of about $8 \times 10^{10}$ cm$^{-2}$. Islands form on top of a ~3 ML thick wetting layer (WL) and tend to flatten during Si capping [Rastelli01]. The observed island density corresponds to a fractional areal coverage of about 70%. For all Si spacer thicknesses investigated here, a planar Si surface is recovered after Si overgrowth of the hut clusters and prior to the growth of the next Ge layer [Denker03-1]. Buried dots influence however the formation and evolution of WL and dots in the subsequent layers, since the Si surface above them presents a lateral strain modulation with amplitude which increases for decreasing $t_{Si}$. As a consequence, the critical thickness for island formation (in first approximation corresponding to the WL thickness) decreases, e.g., from the first Ge layer to second layer. The amount of deposited Ge was therefore properly reduced in the subsequent layers to limit the occurrence of plastically relaxed islands due to the increase of Ge available for island growth [Denker03]. At the same time an attempt was done to keep the amount of deposited Ge as large as possible to keep island density high. Tables SM1 and SM2 summarize the properties of the samples. The Ge volume fraction in the multilayers is estimated as the ratio between the total amount of Ge in the 5 or 11 layers and the total amount of material in the multilayer (amount of Ge plus amount of Si contained in the spacer layers). For the thickness of a ML Ge we have used the value for relaxed Ge (0.1414 nm) and neglected the effect of lattice distortion due to strain. This effect give in fact a negligible contribution to the total thickness of the multilayer in view of the relatively small lattice mismatch between Ge and Si (about 4%) and the small thickness of the Ge layers compared to the Si spacers. Furthermore, actual thickness values may deviate by about $\pm 5\%$ from the nominal values because of growth rate fluctuations from sample to sample. The good stability of the growth rates was verified by comparing nominal thickness values with actual thickness values by transmission electron microscopy (TEM), which was performed on some of the samples studied here. Although the Ge layers on the sample surfaces are oxidized after exposure to air, we decided to take them into account in the estimation of the total multilayer thickness $d$. Their thickness is small compared to the rest of the structure (up to 3% for the thinnest multilayer) and its inclusion/exclusion does not produce any relevant change in the estimation of thermal conductivity values.

For small Si spacer thickness ($t_{Si} = 3$ and 6 nm) islands in subsequent layers are vertically aligned, while for larger $t_{Si}$ islands grow in an uncorrelated fashion. This is illustrated in Fig. SM2, which shows transmission electron microscopy (TEM) images of samples obtained with
same nominal conditions as set #1 (apart from the topmost layer, which is overgrown with Si).

**Fig. SM1:** 3D view of an STM image (scale 56×32×3 nm³) of a Ge hut cluster (from [Rastelli02]). Dots used in this work have the same shape, but have an average size which is about half of the size of the island shown here.

<table>
<thead>
<tr>
<th>$t_{Si}$ (nm)</th>
<th>Nominal amount of Ge in subsequent layers, starting from bottommost layer (ML)</th>
<th>Total multilayer thickness $d$ (nm)</th>
<th>Ge volume fraction in multilayer (%)</th>
<th>Island density on surface layer ($\times10^{10}$ cm⁻²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>6.4, 4.94, 4.58, 4.41, 4.29</td>
<td>15.5</td>
<td>22.5</td>
<td>4.8</td>
</tr>
<tr>
<td>6</td>
<td>6.4, 5.59, 5.47, 5.27, 5.27</td>
<td>28.0</td>
<td>14.2</td>
<td>2.7</td>
</tr>
<tr>
<td>9</td>
<td>6.4, 5.95, 5.83, 5.83, 5.83</td>
<td>40.2</td>
<td>10.5</td>
<td>2.5</td>
</tr>
<tr>
<td>12</td>
<td>6.4, 6.23, 6.23, 6.23, 6.23</td>
<td>52.4</td>
<td>8.4</td>
<td>2.0</td>
</tr>
</tbody>
</table>

**Table SM1:** Sample structure and properties for samples of set #1, containing 5 Ge nanodot layers separated by Si spacer with thickness $t_{Si}$.

<table>
<thead>
<tr>
<th>$t_{Si}$ (nm)</th>
<th>Nominal amount of Ge in subsequent layers, starting from bottommost layer (ML)</th>
<th>Total multilayer thickness $d$ (nm)</th>
<th>Ge volume fraction in multilayer (%)</th>
<th>Island density on surface layer ($\times10^{10}$ cm⁻²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>5.8, 4.6, 4.1, 3.9, 7×(3.7)</td>
<td>36.3</td>
<td>17.3</td>
<td>5.7</td>
</tr>
<tr>
<td>6</td>
<td>5.8, 5.1, 4.9, 4.7, 7×(4.6)</td>
<td>67.5</td>
<td>11.1</td>
<td>1.5</td>
</tr>
<tr>
<td>9</td>
<td>5.8, 5.4, 5.25, 8×(5.2)</td>
<td>98.2</td>
<td>8.3</td>
<td>2.0</td>
</tr>
<tr>
<td>12</td>
<td>5.8, 5.65, 9×(5.6)</td>
<td>129</td>
<td>6.8</td>
<td>1.4</td>
</tr>
</tbody>
</table>

**Table SM2:** Same as table 1 but for samples of set #2, containing 11 Ge nanodot layers.

In order to assess the crystal quality of the grown structures, we have performed extensive cross-sectional TEM investigations of two samples of set #2 (see Fig. SM3 and SM4). Different imaging conditions were used to look for possible dislocations or stacking faults. Normal bright field (BF) TEM images at low magnification of 1 µm long sections at several positions as well as higher magnified BF images of selected section of the layer stack were collected. In total we scanned a 10 µm long section of two prepared sample parts. Dark field TEM images of several selected section of the stacked dot layer were acquired with selected reflexes of the {110} pole. To maximize the contrast, we also tilted the sample into a two-beam condition near the {221} pole. The resulting bright field image is seen in Fig. 1b of the main text. The Ge rich dot layer is clearly resolved as well as the Si spacer. Again larger sections of the sample were systematically imaged and no traces of dislocations resulting from plastic relaxation were found. High resolution TEM (HRTEM) was performed at selected areas in a {110} as well as in a {221} pole alignment of the sample. A typical HRTEM image around one layer of the layer stack is depicted in the inset of Fig. 1b. Clear lattice images with no indication of growth defects were found. Also investigation using Fast Fourier Transformation filtered HRTEM images showed no typical traces of dislocations. Such a study revealed that the multilayers are coherent and no extended defect was found in any of the imaged areas.
Because of the limited sample thickness necessary for HRTEM, TEM alone is not sufficient to rule out the possible presence of dislocations in the structure. We have therefore performed also large scale AFM imaging of the topmost layer of the samples of set #2. It is in fact known that the occurrence of dislocations or other extended defects during the fabrication of the multilayer will locally disrupt the growth of Si or Ge leading to the formation of either large Ge islands or pits/mounds in the Si substrate. For the samples with smallest spacer thickness and smallest cross-plane thermal conductivity (3 and 6 nm) we were not able to detect any of such features in an area as large as 60×60 μm², providing us an estimated upper bound for defect densities of less than 3×10⁴ cm⁻². On the other hand the sample with \( t_{Si} = 12 \) nm shows some large cluster (2 in a 30×30 μm², corresponding to a density of roughly 2×10⁵ cm⁻²) on the surface as shown in Fig. SM5. The density of such clusters (and pits) is higher in the sample with \( t_{Si} = 9 \) nm (1.6×10⁷ cm⁻² large clusters and 1.6×10⁶ cm⁻² pits). Also in this “worst case”, the fractional areal density of the large clusters remains low (below 3%). Since we were not able to detect these defects in TEM because, we are not able to provide details on their nature, but we can ascribe them to the occurrence of plastically relaxed islands in one or more of the layers composing the multilayers. We are confident that by further growth optimization, they can be completely avoided.

We also notice that thermal conductivity measurements show a clear dependence on \( t_{Si} \) while the estimated density of defects fluctuates by 2 orders of magnitudes from sample to sample. This is a further indication that the density of defects remains too low to produce any appreciable effect on thermal conductivity.

**Fig. SM2**: Low resolution TEM image of 5 layers of Ge islands embedded in Si with different interlayer spacer thickness. Arrows in (a) and (b) indicate vertical alignment of islands (from [Denker03-1]).
Fig. SM3: Cross-sectional TEM images of the samples with 11 layers Ge. The bottommost right HRTEM image is taken from the sample with 3 nm Si spacer thickness while the others are from the sample with 12 nm Si spacer thickness. No extended crystal defects are detectable.
**Fig. SM4:** Two HRTEM images of dot layers in two samples with $t_{Si} = 12$ nm (left image) and 6 nm (right image). Both images show clear lattice fringes, signifying that the Ge nanodots are coherently embedded into the surrounding Si matrix.

**Fig. SM5:** Large scale AFM image of the surface of the sample (of set #2) with spacer thickness 12 nm. The inset shows a higher resolution image taken from the surface of the sample with $t_{Si} = 9$ nm in the same set. Large clusters and pits are attributed to the presence of extended defects with low surface density.

**References**

II. Differential $3\omega$ Method

1 Experimental method

The differential $3\omega$ method [Cahill94, Cahill90, Borca01] was employed to determine the cross-plane thermal conductivity of the Ge nanodot multilayers of sample set #2 (see above). In order to carry out the measurements on our semiconducting thin films, an insulating layer was simultaneously deposited on the dot samples and on a reference sample, which consists of Si substrate and MBE-grown Si buffer (no nanodot multilayer). Afterwards, bolometers (metal strips) of about 7 µm width were fabricated on the surfaces by using optical lithography and thermal evaporation of a 5 nm adhesion layer of Cr followed by about 195 nm of Au (Metal evaporation on all samples was performed in the same run to guarantee homogeneous thicknesses).

The capability of the $3\omega$ technique to determine accurately the thermal conductivity of very thin films, such as the dot multilayers considered here, relies on the minimization of various sources of errors and on the correct estimation of the parameters entering in the measurement. In particular, the parasitic thermal resistance of the insulating layer was minimized by scaling down the size of an insulator with relatively high thermal conductivity. While silicon dioxide and silicon nitride layers with thicknesses of some hundred nanometers have been widely used as insulators for the $3\omega$ method [Lee97, Venkatasubramanian00], we have chosen here Al$_2$O$_3$ grown by atomic layer deposition (ALD) [Ritala00, Groner04]. ALD is currently the most used deposition technique for ultrathin electrically insulating layers, because it allows for a high degree of control, homogeneity and reproducibility [Groner04]. It should be noted that a high quality insulating layer is expected to minimize parasitic effects due to the interfacial thermal resistances. All the samples investigated in this work were overgrown by ALD with (29.3±0.5) nm of Al$_2$O$_3$. In this case we estimated the total thermal resistance of the Al$_2$O$_3$ and interfaces (between metal strip and Al$_2$O$_3$ and between Al$_2$O$_3$ and semiconductor) to be of about $2\times10^{-8}$ Km$^2$W$^{-1}$, which is comparable with data reported in Ref. [Behkam05]. This value is also comparable to the thermal resistances of the studied nanodot multilayers (see below). With standard dielectrics of some hundred nanometers thickness, we would have obtained a much lower signal to background ratio and a consequent increase of the uncertainties in the determination of thermal conductivities of the dot multilayers.

We have tested the quality of the insulating layer by measuring the leakage current between two contact pads in a sample without metal strip and found current values which are about 5 orders of magnitude lower than the ones measured in the $3\omega$ device. The inset of Fig. SM6 shows a sketch of the sample structure and metal strip used in our measurements, which were performed at room temperature and in vacuum (pressure of about $10^{-4}$ mbar) with a system developed at the Fraunhofer Institute IPM in Freiburg (Germany).

The temperature coefficient of the resistance of the metal strip was determined by measuring the temperature dependence of the resistance between 300 and 340 K. The sample temperature was set with a Peltier element TEC 3-6 from Thorlabs. An Agilent 33220A signal conditioner was used as a sine wave function generator. The power supplied to the heater was kept constant in all the experiments. The modulation frequency range $f = 40 – 130$ Hz was chosen in order to avoid artifacts at low frequencies due to the finite thickness of the Si substrate and limited at high frequencies by the accuracy of the phase shift measurement that can be obtained by our experimental setup. By repeating measurements on the same samples at different times, we found fluctuations in the results of about 2%, which is lower than other uncertainties and indicate the good reproducibility of the results with the used system.

Since the accurate knowledge of the width of the metal strips used in different samples is crucial for the determination of the thermal conductivity values, we have measured the average strip width of all the samples by 3 independent methods: atomic force microscopy.
(AFM), optical microscopy and electric resistance measurements. The latter allows us to estimate the width of the strip on a sample relative to the strip width on another sample under the assumption that the metal thickness is constant. All methods provide values which are well compatible with each other.

Figure SM6 shows the raw $3\omega$ data (real part of the temperature oscillations vs logarithm of the heater frequency) from the samples of set #2 (see section I). Since the differences in the strip widths are small (see Table SM4), Fig. SM6 demonstrates that the presence of the dot multilayer produces a frequency independent increase in the in-phase $\Delta T$ with respect to the reference sample, which contains no dots. In spite of the limited thickness of the multilayers (from about 35 to 130 nm), the extra temperature rise, and hence the extra thermal resistance produced by the Ge layers is always visible, reproducible and large enough to be precisely quantified. Furthermore, the fact that all the data cluster together indicates that the thermal resistances are similar for all the investigated dot samples. A precise determination of the thermal resistance of the different multilayers requires taking into account differences in strip width, as explained below.

![Figure SM6: Raw $3\omega$ data at room temperature for the samples containing 11 layers of Ge dots separated by 3, 6, 9 and 12 nm thick Si spacers, respectively. The measurement of a sample without Ge (Reference) is also shown. Inset: schematic illustration of the structures for $3\omega$ measurement along with the top view of the metal strip, which has a width of $2b$.](image)

### 2 Data Analysis

Data reduction for the $3\omega$ method relies on heat conduction models, the applicability and accuracy of which is often restricted by specific constraints [Cahill90, Borca01]. In this work, a one-dimensional heat conduction analysis is applied to determine the multilayer thermal conductivity. In fact, the SiGe multilayer thickness is always more than one order of magnitude smaller than the width of the metal line, leading to negligible lateral heat spreading effects in the SiGe heterostructure [Borca01, Koh09]. In addition, according to previous literature reports, the thermal conductivity of the Si substrate is expected to be much larger than the one of the Si/Ge multilayers, justifying a simple thermal resistance assumption for the dielectric and the dot multilayer [Cahill94, Liu01].

The temperature drop across the thin film can be therefore obtained, within the framework of the differential $3\omega$ technique, from the difference between the total temperature rise measured in the samples with nanodots and the temperature drop measured across a reference sample without the thin film of interest. This approach is based on the assumption that the strip width
is exactly the same for the reference and the investigated samples. In reality, this requirement is met within certain limits, which depend on the accuracy and reproducibility of the lithographic steps used to define the metal strips. We accounted for the sample-to-sample strip width fluctuations as follows.

The analytical expression for the real part of the temperature rise developed in the reference samples, \( \Delta T_1 \), is [Cahill94, Borca01]:

\[
\Delta T_1 = \frac{P_l}{\pi \kappa_S} \left\{ \frac{1}{2} \ln\left( \frac{\kappa_S}{C \rho b_{ref}^2} \right) + \eta - \frac{1}{2} \ln(2\omega) \right\} + \frac{P_l}{2b_{ref}} R_D = \Delta T_{\text{sub}}(b_{\text{ref}}, \omega) + \frac{P_l}{2b_{\text{ref}}} R_D \tag{1}
\]

where \( \omega \) is the angular frequency of the heating current, \( P_l \) is the average power per unit length, \( 2b_{\text{ref}} \) is the width of the metal line for the reference sample, \( \eta \) is a constant. Here \( K_S, C \) and \( \rho \) are the thermal conductivity, specific heat, and density of the Si substrate, respectively [Jacquot02]. The cross-plane heat transport in the dielectric adds a frequency-independent component to the total thermal response as described by the thermal resistance \( R_D \). Likewise, the experimental temperature rise of the heater on the dot sample, \( \Delta T_2 \), can be written as:

\[
\Delta T_2 = \frac{P_l}{\pi \kappa_S} \left\{ \frac{1}{2} \ln\left( \frac{\kappa_S}{C \rho b_{\text{ND}}^2} \right) + \eta - \frac{1}{2} \ln(2\omega) \right\} + \frac{P_l}{2b_{\text{ND}}} R_{\text{TOT}} = \Delta T_{\text{sub}}(b_{\text{ND}}, \omega) + \frac{P_l}{2b_{\text{ND}}} \left( R_D + \frac{t_{\text{ND}}}{\kappa_{\text{ND}}} \right) \tag{2}
\]

where \( 2b_{\text{ND}} \) is the strip width for the nanodot multilayer sample, \( t_{\text{ND}} \) and \( \kappa_{\text{ND}} \) are the multilayer thickness and thermal conductivity, respectively. Since the dielectric was deposited on the two samples during the same deposition run, the insulator thermal resistance can be assumed as a constant. The thermal conductivity of the multilayer can be therefore determined from Eqs. 1 and 2 as follows:

\[
\kappa_{\text{ND}} = \frac{P_l}{2b_{\text{ND}}} \left[ \frac{t_{\text{ND}}}{\Delta T_2 - \Delta T_{\text{sub}}(b_{\text{ND}}, \omega) - \frac{b_{\text{ref}}}{b_{\text{ND}}} (\Delta T_1 - \Delta T_{\text{sub}}) \right] \tag{3}
\]

The resulting multilayer thermal conductivities obtained for the sample set #2 (see section I above) are summarized in Tab. SM3. For each sample these results were obtained by averaging the data over the frequency range of 40-130 Hz and uncertainties were estimated as described in section II.3 below. Although the raw data displayed in Fig. SM6 do not show any clear trend, the total thermal resistance of the multilayers obtained after taking into account differences in strip widths shows a slight increase with increasing spacer thickness. We tentatively attribute this increase to the fact that the absolute amount of Ge is increasing with increasing \( t_{\text{Si}} \) (see 2\textsuperscript{nd} column of Table SM2). In fact, after normalizing the thermal resistance for the total amount of Ge, we obtain a rather constant value (see Fig. 2c of the main text).

It is worth noting that the aforementioned approach is nearly insensitive to the substrate thermal conductivity (within 3\% by varying the thermal conductivity of the Si substrate from 130 to 150 W/m-K), and any additional contributions from thermal boundary resistances is minimized by comparing two measurements performed on samples having nominally identical metal-dielectric and dielectric-thin film interfaces.

For the sake of completeness, we have also attempted to measure the contribution to the thermal resistance of the system provided by a single layer of Ge dots on the sample surface (the same sample shown in Fig. 1c of the main text, which consists of 5.8 ML Ge grown on a 100 nm Si buffer layer). By comparing the result with that obtained on the reference sample (with no dots) we obtained values which are compatible within the uncertainties: our present errors do not allow us to determine the thermal resistance of a single layer of Ge dots relative to a bare Si interface.
Furthermore we have compared the reference structure with a bare Si substrate (with no MBE grown Si buffer) and found compatible results, indicating that the Si buffer has thermal properties which are indistinguishable from those of the Si substrate within the uncertainties of our measurements.

<table>
<thead>
<tr>
<th>Thickness of Si Spacer $t_{Si}$ (nm)</th>
<th>Thermal resistance (K·m²/W)×10⁻⁹</th>
<th>Thermal conductivity (W/m/K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>27±7</td>
<td>1.4±0.5±0.3</td>
</tr>
<tr>
<td>6</td>
<td>30±8</td>
<td>2.3±0.8±0.5</td>
</tr>
<tr>
<td>9</td>
<td>34±8</td>
<td>2.9±0.9±0.6</td>
</tr>
<tr>
<td>12</td>
<td>35±8</td>
<td>3.7±1±0.7</td>
</tr>
</tbody>
</table>

Table SM3: Thermal conductivity results for the sample set #2 (11 Ge layers). The quoted uncertainties are evaluated as described below and represent the limits of a confidence interval with 68.3% confidence level.

### 3 Uncertainty evaluation on thermal conductivity data: error propagation by Monte Carlo simulation

Equation 3, which is used for the determination of the thermal conductivity, depends on several experimentally measured quantities (with their own uncertainties) in a non linear fashion. Furthermore, since uncertainties are relatively large when measuring very thin films, common error propagation approaches are no longer accurate. To address this issue, we performed a simple Monte Carlo simulation to propagate the uncertainties. In this approach we randomly generated all the quantities entering in Eqs. 1-3 according to their measured value, uncertainty, and expected statistical distribution (see Tab. SM4). From each set of random values we obtained, from Eq. 3, the corresponding value of $\kappa_{ND}$. By iterating the simulation step many times (here 20000 for each value of frequency) and by plotting the results in form of a histogram we obtained an estimate for the probability density distribution for the thermal conductivity (“bootstrap method” [NumRec]).

As an example, Fig. SM7 shows the histogram (properly normalized) for the sample with 3 nm Si spacer. Analogous results were obtained for all the other samples. As expected, the simulated probability density distribution of the thermal conductivity does not follow a normal distribution and is slightly asymmetric. To estimate the uncertainty on the measurements, we defined a confidence interval containing 68.3% (confidence level, CL) of the events around the measured value, as depicted in Fig. SM7. The error bars shown in Tab. SM3 and in Fig. 2 of the manuscript represent the upper and lower limits of such interval. The fact that the error bars are not symmetric is a consequence of the skewed probability density distribution.

<table>
<thead>
<tr>
<th>$t_{Si}$ (nm)</th>
<th>Total thickness $d$ (nm)</th>
<th>Thickness Uncertainty (%)</th>
<th>Strip width (µm)</th>
<th>Strip width uncertainty (µm)</th>
<th>Power (W/m)</th>
<th>Power uncertainty (%)</th>
<th>$T$ uncertainty (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>36.3</td>
<td>5</td>
<td>6.2</td>
<td>0.5</td>
<td>22</td>
<td>2.8</td>
<td>4</td>
</tr>
<tr>
<td>6</td>
<td>67.5</td>
<td>5</td>
<td>7.5</td>
<td>0.5</td>
<td>22</td>
<td>2.8</td>
<td>4</td>
</tr>
<tr>
<td>9</td>
<td>98.2</td>
<td>5</td>
<td>7.3</td>
<td>0.5</td>
<td>22</td>
<td>2.8</td>
<td>4</td>
</tr>
<tr>
<td>12</td>
<td>129</td>
<td>5</td>
<td>7.2</td>
<td>0.5</td>
<td>22</td>
<td>2.8</td>
<td>4</td>
</tr>
<tr>
<td>Reference</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table SM4: Inputs and their density distributions employed in the Monte Carlo uncertainty evaluation.
Figure SM7: Estimated probability density distribution for the thermal conductivity of the sample with 11 layers of Ge dots and a 3 nm-thick Si spacer. The graph, obtained by Monte Carlo simulation, is used to propagate the errors on the experimentally measured quantities which enter into the model Eq. 3. The white area around the measured value (blue line) contains in total the 68.3% (CL) of the counts of the histogram.

Although this method is usually much better than the usual error propagation, it remains nevertheless approximate, because the shape of the distribution around the measured parameters is assumed to be the same of that around the true ones [NumRec].

4 Measurement validation

In this work the differential $3\omega$ method has been used as a means to measure the cross-plane thermal transport in layers as thin as 35 nm. To provide an independent test on the accuracy of the method, we measured the thermal conductivity of a well-known standard sample. We mimic the sample structure by replacing the dot multilayer with a 50 nm-thick a-SiO$_2$ layer, for which both the thermal conductivity and the thickness are comparable to the multilayer samples [Lee97].

Figure SM8(a) shows the frequency dependence of the in-phase temperature oscillations for a reference Si substrate sample and for the 50 nm-thick SiO$_2$ film, which was thermally grown on Si in the same run as the sample used for the validation of the HPTR method. The two samples were overgrown by about 30 nm of Al$_2$O$_3$ as for the dot multilayers, while the strip widths were 7.3 $\mu$m and 5.9 $\mu$m for the reference and the SiO$_2$ sample, respectively. The thermal response of the sample with the extra SiO$_2$ layer has an additional frequency-independent component with respect to the cross-plane heat transport in the substrate.

The thermal conductivity of the 50 nm-thick SiO$_2$ layer is then determined by using the aforementioned method, yielding a value of:

$$\kappa_{\text{SiO}_2} = 1.5^{+0.5}_{-0.3} \text{ W m}^{-1} \text{ K}^{-1}$$

(see Fig. SM8(b)). This result is in very good agreement with the value of $\kappa$(thermal oxide) = 1.47 W/m-K quoted in Ref. [Chien08] and is fully consistent with the result of the HPTR method reported in this work (see Sec. III.2 below). This test provides further evidence that the thermal boundary resistance plays a negligible role in our measurements.
There are indeed several reports where the thermal conductivity of thin film layers (as low as tens of nanometers) were inferred from the temperature rise difference between the measured sample and the calculated value of the bare substrate [Lee97, Chien08, Yamane02]. It was found that the measured thermal conductivity was a function of the layer thickness. For instance, the observed thermal conductivity in SiO₂ films of about 50 nm was about a factor of two lower than the “bulk” value [Lee97, Chien08]. In some works these findings have been analyzed in terms of a parasitic effect due to the presence of interface thermal resistances [Chien08]. This systematic decrease was not observed here, and can be ruled out because an apparent thermal conductivity of 0.74 W/m K, as obtained from Ref. [Chien08], lays in a 0.2‰ probability interval (see Fig. SM8). The agreement of our findings with bulk data demonstrates the effectiveness of the differential 3ω method to determine the intrinsic thermal conductivity of thin films. The thermal boundary resistance contribution, albeit present, is indeed minimized by comparing the sample of interest with a reference specimen which contains all the layers (substrate, dielectric, metal strips) but the interesting one (SiO₂ or Ge/Si multilayer).

References

III. Thermal Parameters Estimation Using Heterodyne Picosecond Thermoreflectance

1 Thermal Transport Model

We developed a theoretical model to calculate the thermal response of a multi-layered structure examined with the HPTR technique.

First we calculate the thermal response to a single pulse. The thermal transport model is based on the Fourier heat equation and the following four hypotheses:

i. The laser pulse trains are assumed to be temporal Dirac combs.
ii. The laser profile is assumed to be Gaussian and the material properties to be isotropic with 3D propagation in cylindrical symmetry.
iii. The temperature field in the metal film is uniform in the light penetration depth, thus the metal layer is split into two layers where the 10 first nanometers are assumed to be an effective transducer. These two layers have obviously the same thermal properties and there is no contact resistance between them.
iv. The Si substrate is thermally thick and taken as a semi-infinite layer.

The Fourier heat equation is solved using numerical Laplace-Hankel integrals transform; by the “Thermal Quadrupole Model” [Maillet00].

We assume the relative reflectivity changes of the material to be proportional to the temperature variation:

\[ \frac{\Delta R(t)}{R} \propto \Delta T(t) \]

Since the measured signal \( S(t) \) is induced by a pulsed laser one has to take into account the cumulative effect generated by the previous pulses. It is expressed as follows [Dilhaire09]:

\[ S(t) \propto \sum_{n=0}^{N} \sum_{q=0}^{+\infty} \Delta T \left( qT_p + n\Delta T \right) \delta \left( t - nT_p - n\Delta T \right) \]

(4)

Where \( N \) is the number of time steps in the experiment, \( T_p \) is the pump repetition period and \( q \) is the index of each previous pump pulse. The identification of thermal parameters is performed with Eq (4).

2 Validation of the method on a 50nm SiO\(_2\) layer

To check our identification procedure, we carried out HPTR measurements on a 50nm thermally grown SiO\(_2\) sample caped with aluminum. The description of the sample is given in the figure below:
We start our optimization process 100ps after the coincidence between pump and probe pulses to safely assume that the temperature is homogeneous along the optical absorption depth in the metal film and to avoid non-thermal effects such as acoustic echoes.

We kept only one free parameter: the thermal conductivity $\kappa$ of SiO$_2$ layer. As in Ref [Costescu03], the interface thermal resistances are included in the total thermal resistance of the SiO$_2$ layer. The aluminum thickness was measured by a Tencor Alpha-Step 500 profiler. The other parameters were fixed with the literature values.

As we found a very low sensitivity to the Si substrate properties only the uncertainty on the metal layer properties was taken into account for the error analysis. To obtain the extreme values of the thermal conductivity, we have varied the quantity $(\rho C_p d)_{Al}$ by 10%. ($\rho$ is the density, $C_p$ the specific heat and $d$ the thickness of the aluminum layer). Thus we obtain the following value for the thermal conductivity:

$$\kappa_{SiO_2} = 1.33 \pm 0.15 \text{ W.m}^{-1}\text{.K}^{-1}$$

This result in excellent agreement with the values found in previous reports [Costescu03, Cahill00] and with the $3\omega$ results on the same material (see Sec. II.4 above). It also indicates that the metal/SiO$_2$ and SiO$_2$/Si interface resistances, which are included in the thermal resistance of the 50-nm thick SiO$_2$ layer, do not produce significant effects on the intrinsic value of $\kappa_{SiO_2}$, in agreement with the findings in Ref. [Costescu03]. On the other hand, for the measurements on the nanodot samples, we have explicitly identified also the metal/semiconductor contact resistance, as discussed in the following section.

### 3 Sample description and modeling

We studied the two sets of samples described in Sec. I. Here we present our model for thermal parameter identification in these 8 samples.

In order to address the uncertainty due to the thermal contact resistance between the metal layer and the multilayer, the sample is modeled as an interface resistance ($R_{int}$) on top of an effective layer with thickness $d$ and thermal conductivity $\kappa_{ND}$, the total nanodot-multilayer thermal resistance $R_k$ is given by:

$$R_k = R_{int} + d / \kappa_{ND} \quad (5)$$

Results for the identified values of ($R_{int}$) and ($\kappa_{ND}$) are given in the Sec III.5.
The total thickness, $d$, reported in Tab. SM1 and SM2 of Sec. I above is then used to determine the equivalent thermal conductivity using equation 5.

We would like to point out that the interface thermal resistance ($R_{int}$) and the multilayer thermal resistance ($d / \kappa_{ND}$) are actually correlated for time scales larger than 5000 ps as can be seen on Figure SM11.

We define the sensitivity function for the parameter $P$ as:

$$Sens(P) = \frac{\partial \Delta S_{calculated}(t)}{\partial P} \quad (6)$$

where $\Delta S_{calculated}(t)$ is the calculated temperature given by the thermal model.

Figure SM10: Sample description of the multilayered structure where $t_{Si}=3\text{nm}, 6\text{nm}, 9\text{nm}$ and $12\text{nm}$.

Figure SM11: The interface thermal resistance and the multilayer thermal resistance sensitivity functions for a sample with 5 Ge layers.
The picosecond time resolution of our experiment allows us to identify the interface thermal resistance in the first hundreds of picoseconds of the thermal decay. HPTR measures the thermal decay over 4 time decades, this is where its great strength resides.

4 HPTR measurements

We performed HPTR measurements on 8 samples described in the article and in Sec. I. Figure SM12 shows the raw thermoreflectance responses normalized with the same experimental conditions (10× magnification objective, 50mW pump power @780nm wavelength, 15mW probe power @810nm wavelength).

![Normalized ΔR/R](image)

**Figure SM12:** The relative change of reflectivity of the 4 samples (3, 6, 9, 12 nm Si spacer) over 13ns with 1ps temporal resolution (left: sample set #1; right: sample set #2 – see Sec. I for details on the samples).

The relative changes of reflectivity are proportional to the sample surface temperature variation. The thermal decay is linked to the thermal properties of the samples.

5 Parameter identification

5.1 Method

Thermal properties are obtained by fitting the experimental signal (∆S<sub>exp</sub>(t)) with the temperature calculation (∆S<sub>calculated</sub>(t)). The optimization is based on the least-square method where we minimize an objective function (f<sub>obj</sub>) defined by the difference between the measured and calculated signals:

\[
f_{obj}(P_1,\ldots,P_n) = \sum_i (\Delta S_{exp}(t_i) - \Delta S_{calculated}(t_i, P_1,\ldots,P_n))^2
\]

For minimization we use a standard Levenberg-Marquardt algorithm [Beck76].
We performed sensitivity studies with respect to all thermophysical parameters. The results show a dominant sensitivity to the aluminum parameters \((\rho C_p d)_{Al}\) and to the thermal parameters of the multilayer. In our first fitting attempt, where these parameters were free, we noticed a correlation between them. In subsequent attempts we fixed the aluminum parameters \((\rho C_p d)_{Al}\). The specific heat was taken from literature and the thickness determined by AFM measurement. The Si substrate and buffer parameters were fixed with the literature values (see table SM5). It is important to note that they do not affect the surface temperature as they are deeply buried and their sensitivity functions are very small. Furthermore, as noted in Sec. II.2, 3o measurements did not show any significant difference between the MBE-grown Si material and the bare Si substrate.

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Si Thermal conductivity</td>
<td>130 W/m/K</td>
</tr>
<tr>
<td>Si Specific heat</td>
<td>1.624 \times 10^6 J/K/m^3</td>
</tr>
</tbody>
</table>

**Table SM5:** Values of thermal properties used for the Si-substrate and Si buffer layer.

The pump and probe beam waists were measured with a Beam Master™ laser profiler, they are respectively equal to 6.5±0.5µm and 5.5±0.5µm with a M² factor equal to 1.2.

To estimate the uncertainties we report the extreme values found when all the parameters are fixed and obtained from literature (heat capacity of aluminum, thermal conductivity of silicon) with a variation of ±10%. For the aluminum thickness (measured by AFM profilometry) the uncertainty is caused by the residual roughness. Uncertainties associated to these values are detailed in the section 6.

Figure SM13 shows the residual signal \((\Delta S_{\text{exp}}(t) - \Delta S_{\text{calculated}}(t,P_1,...,P_n))\) obtained on the sample \(t_{Si}=3\) nm and 5 Ge layers. The residual error is less than 0.5%.

**Figure SM13:** Difference between experimental and optimized signals for the sample with \(t_{Si}=3\) nm and 5 Ge layers.
5.2 RESULTS

As the multilayer is modeled by an interface resistance and an effective layer, thus the total thermal resistance is given by:

\[ R_k = R_{\text{int}} + \frac{d}{\kappa_{ND}} \]

with \( d \) reported in Tables SM1 and SM2. Results for the identified values of \((R_{\text{int}})\) and \((\kappa_{ND})\) for 5 and 11 Ge layers are reported in Tables SM6 and SM7. We also calculated the total resistance \((R_k)\).

<table>
<thead>
<tr>
<th>( t_0 ) (nm)</th>
<th>( d ) Total thickness (nm)</th>
<th>( R_{\text{int}} ) Interface thermal resistance (m²K/W) ( \times 10^{-9} )</th>
<th>( \kappa_{ND} ) Thermal conductivity (W/m-K)</th>
<th>( R_k ) Total thermal resistance (m²K/W) ( \times 10^{-9} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>15.5</td>
<td>1.0( \pm )0.2</td>
<td>0.9( \pm )0.1</td>
<td>18.6( \pm )2.0</td>
</tr>
<tr>
<td>6</td>
<td>28.0</td>
<td>2.2( \pm )0.5</td>
<td>1.4( \pm )0.2</td>
<td>21.7( \pm )3.3</td>
</tr>
<tr>
<td>9</td>
<td>40.2</td>
<td>2.4( \pm )0.5</td>
<td>2.1( \pm )0.2</td>
<td>21.6( \pm )2.3</td>
</tr>
<tr>
<td>12</td>
<td>52.4</td>
<td>2.4( \pm )0.5</td>
<td>2.6( \pm )0.3</td>
<td>22.2( \pm )2.8</td>
</tr>
</tbody>
</table>

Table SM6: identified parameters for the samples of set #1 (see table SM1 for details) in the case of a thermal interface \( R_{\text{int}} \) and an effective layer.

<table>
<thead>
<tr>
<th>( t_0 ) (nm)</th>
<th>( d ) Total thickness (nm)</th>
<th>( R_{\text{int}} ) Interface thermal resistance (m²K/W) ( \times 10^{-9} )</th>
<th>( \kappa_{ND} ) Thermal conductivity (W/m-K)</th>
<th>( R_k ) Total thermal resistance (m²K/W) ( \times 10^{-9} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>36.3</td>
<td>2.8( \pm )0.4</td>
<td>1.3( \pm )0.2</td>
<td>30.5( \pm )4.7</td>
</tr>
<tr>
<td>6</td>
<td>67.5</td>
<td>3.7( \pm )0.6</td>
<td>2.1( \pm )0.2</td>
<td>35.9( \pm )3.7</td>
</tr>
<tr>
<td>9</td>
<td>98.2</td>
<td>4.0( \pm )0.6</td>
<td>2.7( \pm )0.3</td>
<td>40.5( \pm )4.6</td>
</tr>
<tr>
<td>12</td>
<td>129</td>
<td>3.7( \pm )0.6</td>
<td>3.4( \pm )0.5</td>
<td>42.0( \pm )6.1</td>
</tr>
</tbody>
</table>

Table SM7: identified parameters for the samples of set #2 (see table SM2 for details) in the case of a thermal interface \( R_{\text{int}} \) and an effective layer.

For each case we found the interface resistance of the Al/multilayer \( (R_{\text{int}}) \) to be small with respect to the thermal resistance of the multilayer \( (d/\kappa_{ND}) \). We also observe that the interface resistance values fluctuate from sample to sample and they tend to be larger for the sample set with 11 Ge layers. Although the origin of such differences is at present unclear, we qualitatively ascribe them to the fact that the two sample sets were grown at different times and also the HF cleaning and metallization were done in different runs. A more systematic investigation will be required to find the impact of different processing steps on the contact resistance.
6 Error analysis

The thermal conductivity uncertainty estimation requires knowledge of the parameter uncertainties used in the model such as aluminum thickness and specific heat, and the Si thermal conductivity.

In the following paragraphs, we give variations of the thermal resistance induced by maximum permissible measurement errors. Results are detailed for the sample with \( t_{\text{Si}} = 12 \) nm and 11 Ge layers corresponding to a total multilayer thickness of 129 nm.

**EFFECT OF ALUMINUM LAYER UNCERTAINTIES**

The thermal response is very sensitive to the top metal layer heat capacity expressed by the product \((\rho C_p d)_{Al}\).

\[
(\rho C_p d)_{Al} = 2.42 \times 10^6 \text{J.K}^{-1} \text{m}^{-3} \times 42 \text{nm}
\]

Although the value of specific heat of aluminum is well known from literature, an oxide layer of Al\(_2\)O\(_3\) is present at the surface and could influence the heat capacity of the top layer [Cahill04]. Moreover, to take into account the aluminum thickness uncertainty, we applied a relative change of \(\pm 10\%\) to the product \((\rho C_p d)_{Al}\). We thus identified the following thermal values:

<table>
<thead>
<tr>
<th>((\rho C_p d)_{Al}) (\pm 10%)</th>
<th>(R_{\text{int}}) Interface thermal resistance ((\text{m}^2\text{K}/\text{W}) \times 10^{-9})</th>
<th>(\kappa_{\text{ML}}) Thermal conductivity ((\text{W/m-K}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>((\rho C_p d)_{Al}) (+10%)</td>
<td>3.4</td>
<td>3.8</td>
</tr>
<tr>
<td>((\rho C_p d)_{Al})</td>
<td>3.7</td>
<td>3.4</td>
</tr>
<tr>
<td>((\rho C_p d)_{Al}) (-10%)</td>
<td>4.4</td>
<td>2.9</td>
</tr>
</tbody>
</table>

Table SM8: Effect of top layer heat capacity variation on the identified interface resistance and thermal conductivity. The error bars correspond to a relative uncertainty of about 15\% for the thermal conductivity.

**EFFECT OF SILICON THERMAL CONDUCTIVITY AND PUMP/PROBE RADI INUS**

The classical value for Si thermal conductivity found in literature is \(\kappa_{\text{Si}} 130\) W/m/K. Using the same method as described below, we also applied \(\pm 10\%\) variation. Results are summarized in table SM9.

<table>
<thead>
<tr>
<th>Si thermal conductivity ((\text{W/m-K}))</th>
<th>(R_{\text{int}}) Interface thermal resistance ((\text{m}^2\text{K}/\text{W}) \times 10^{-9})</th>
<th>(\kappa_{\text{ML}}) Thermal conductivity ((\text{W/m-K}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>143 ((+10%))</td>
<td>3.73</td>
<td>3.36</td>
</tr>
<tr>
<td>(\kappa_{\text{Si}} ,(130))</td>
<td>3.71</td>
<td>3.37</td>
</tr>
<tr>
<td>117 ((-10%))</td>
<td>3.69</td>
<td>3.37</td>
</tr>
</tbody>
</table>

Table SM9: Effect Silicon properties on the identified interface resistance and thermal conductivity.

The very low sensitivity, less than 1\%, is essentially caused by the depth and the high conductivity of the substrate.
We have considered the uncertainties on pump and probe radii as well; the response does not show a significant sensitivity to these parameters (about 1%) as only the amplitude of the signal is affected.

Hence, we can consider that the main source of uncertainties comes from the aluminum properties.

References

IV. Comparison of phonon mean free paths between planar superlattices and nanodot multilayers

The theoretical atomistic calculation permits to elucidate the reason for the larger thermal conductivities reported on single crystalline planar superlattices [Lee97], as compared to those we have measured on nanodot multilayers. The main difference between the computed thermal conductivity of planar superlattices and nanodot multilayers comes from the contribution of phonon frequencies between about 1 and 20 THz, where the added roughness of the dot layers reduces their mean free path with respect to the planar superlattice case. Phonons in that frequency range contribute more than half of the total thermal conductivity, and therefore the thermal conductivity of the planar superlattices is considerably larger than that of the nanodot multilayers. At higher frequencies, above 30 THz, the phonon wavelengths are already quite short, so the mean free paths are determined by atomic level disorder due to the alloy structure of the barriers, which is similar in the two systems (see Fig. SM14). Therefore, this higher frequency range contributes more or less equally in the two different systems.

The computed phonon mean free paths (MFP) are shown in the figure below, for the case of 6.5 nm spacing between barriers.


Figure SM14: AGF computed phonon mean free paths at 300K for Si_{0.5}Ge_{0.5} alloy (red), flat barrier superlattice (black), and nanodot multilayer (green.)