

Thermal boundary conductance under large temperature discontinuities of ultrathin Pb(111) films on Si(111)

Cite as: Appl. Phys. Lett. **127**, 172202 (2025); doi: [10.1063/5.0291617](https://doi.org/10.1063/5.0291617)

Submitted: 18 July 2025 · Accepted: 11 September 2025 ·

Published Online: 28 October 2025



Christian Brand,^{1,2,a)} Tobias Witte,¹ Mohammad Tajik,¹ Jonas D. Fortmann,¹ Birk Finke,¹ Herbert Pfnür,² Christoph Tegenkamp,^{2,3} and Michael Horn-von Hoegen^{1,4}

AFFILIATIONS

¹Faculty of Physics, University of Duisburg-Essen, 47057 Duisburg, Germany

²Institut für Festkörperphysik, Leibniz Universität Hannover, 30167 Hannover, Germany

³Institut für Physik, Technische Universität Chemnitz, 09126 Chemnitz, Germany

⁴Center for Nanointegration (CENIDE), University of Duisburg-Essen, 47057 Duisburg, Germany

^{a)}Author to whom correspondence should be addressed: christian.brand@uni-due.de

ABSTRACT

Heat transfer is a critical aspect of modern electronics, and a deeper understanding of the underlying physics is essential for building faster, smaller, and more powerful devices with improved performance and efficiency. In such nanoscale structures, the heat transfer between two materials is limited by the finite thermal boundary conductance across their interface. Using ultrafast electron diffraction under grazing incidence we investigated the heat transfer from ultrathin epitaxial Pb films to an Si(111) substrate under strong nonequilibrium conditions. Upon applying an intense femtosecond laser pulse, the 5–7 ML thin Pb film undergoes rapid heating by 10–120 K while the Si substrate remains cold at ≈ 10 K. At such large temperature discontinuities, a significantly faster cooling is observed for more strongly excited Pb films. The decrease in the corresponding cooling time constant is explained by variations in thermal boundary conductance, interpreted within the framework of the diffuse mismatch model. The thermal boundary conductance is reduced by more than a factor of three in comparison with Pb films grown on H-terminated substrates, underscoring the importance of substrate, heterofilm, and interface morphologies.

Published under an exclusive license by AIP Publishing. <https://doi.org/10.1063/5.0291617>

As the spatial dimensions of modern electronic devices are down-scaled to the nanometer regime, heat dissipation from an electronically active medium across an interface toward the substrate is no longer governed by the thermal conductivity κ of the bulk but rather limited by the thermal boundary conductance G of the interface.^{1–3} The discontinuity of elastic properties, such as sound velocity and phonon density of states (PDOS), between the film and substrate lead to additional thermal resistance, which is accompanied by a discontinuity in temperature ΔT at the interface.⁴ For films thinner than the Kapitza length κ/G ($\approx 10 \mu\text{m}$ for Pb/Si at 100 K), heat transfer is governed by interfacial conductance, whereas for thicker films the bulk thermal conductivity dominates.

Usually, G is determined by techniques such as time domain^{1,5–9} and frequency domain^{10–12} thermoreflectance or 3ω method^{5,13,14} under conditions where the equilibrium between film and substrate is

only slightly distorted. Thus, the temperature difference ΔT between film and substrate is small compared to the temperature of the substrate. However, such a situation might be untypical for technological applications, e.g., electronic devices in which heat dissipation is strongly localized at the nanoscale, and large temperature discontinuities may occur at the interfaces with heat fluxes reaching up to $100 \text{ kW}/\text{cm}^2$.¹⁵ Under such conditions, the heat transfer across interfaces is no longer defined by a constant G but becomes temperature-dependent. Cases with large temperature discontinuity at the interface, however, are not accessible using conventional experimental techniques such as the thermoreflectance or the 3ω method.

Here, we used ultrafast reflection high energy electron diffraction (URHEED) as a thermometer to follow the transient temperature evolution of the heterofilm upon impulsive excitation by a femtosecond laser pulse^{16–21} [cf. Fig. 1(a)]. RHEED at grazing incidence ensured

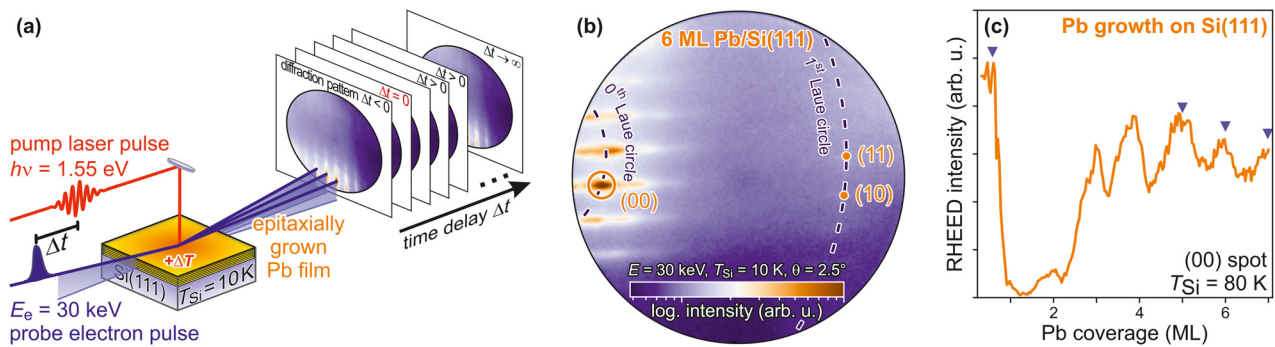


FIG. 1. URHEED experiment: (a) Schematic of the laser pump–electron probe setup for the Pb/Si(111) heterosystem. (b) RHEED pattern at an electron energy of 30 keV taken from a 6 ML thin epitaxial Pb(111) film grown on Si(111). (c) Thickness calibration through layer-by-layer RHEED intensity oscillations of the (00) spot as function of Pb coverage during growth by MBE. The blue triangles indicate the positions where the shutter of the evaporator was opened and closed, i.e., at 1/3 ML Pb coverage (Si(111)- $\beta(\sqrt{3} \times \sqrt{3})R30^\circ$ -Pb wetting layer), and at 5, 6, and 7 ML Pb coverage, respectively.

surface sensitivity,²² while large intensity changes upon changes of temperature on the order of $I/I_0 \approx 0.5\%$ per Kelvin were observed. We employed the Debye–Waller effect to determine the transient change in temperature $\Delta T(\Delta t)$ from the drop of diffraction spot intensity $\Delta I(\Delta t)$.^{23,24}

Here, we nicely turned a seeming disadvantage into an advantage: the URHEED technique requires high excitation density, i.e., large temperature rises ΔT on the order of 10–120 K, subsequent to a short laser pulse to achieve a sufficient signal-to-noise ratio; this requirement is ultimately constrained by the point-to-point stability and longtime drift of the femtosecond pulses from the laser amplifier. This allowed us to determine the thermal boundary conductance under extreme nonequilibrium conditions at large temperature discontinuities $\Delta T \gg T_{\text{Si}}$, i.e., at a Si substrate temperature of $T_{\text{Si}} = 10$ K, the temperature rise of the film exceeded the substrate temperature by more than ten times at the highest incident laser pulse energy used.

All experiments were performed under ultra-high vacuum (UHV) conditions at a base pressure of 4×10^{-10} mbar. The Si(111) substrate ($\pm 0.1^\circ$, 380 μm , 0.6–1 Ωcm , n -doped with $6\text{--}10 \times 10^{15}$ P atoms per cm^3 , Virginia Semiconductor) was mounted on a cryostat cooled by liquid He. The substrate was prepared by degassing at 600 $^\circ\text{C}$ for several hours followed by flash-annealing at 1250 $^\circ\text{C}$ to desorb the native oxide and prepare a clean (7×7) -reconstructed surface. The Pb film was grown by molecular beam epitaxy (MBE) from an indirectly heated Knudsen-type ceramic crucible.²⁵ As a template for film deposition, a Si(111)- $\beta(\sqrt{3} \times \sqrt{3})R30^\circ$ -Pb reconstruction was prepared through Pb deposition and followed by annealing for 20 s, both at 600 $^\circ\text{C}$. Subsequently, using the kinetic pathway, continuous epitaxial Pb films of $d = 5\text{--}7$ ML thickness (1 monolayer (ML) Pb(111) $d = 2.86$ \AA , 1.02×10^{15} $\text{\AA}/\text{cm}^2$) were grown at low sample temperature ≈ 80 K [cf. RHEED pattern in Fig. 1(b)], thus avoiding islanding of the Pb films,²⁶ and resulting in a smooth surface and high quality of the Pb films as proven by *in situ* low energy electron diffraction. The film thickness was calibrated through RHEED intensity oscillations of the (00) spot during layer-by-layer growth^{27,28} as shown in Fig. 1(c).

The ultrathin Pb films were impulsively excited by 80 fs laser pulses at a photon energy of 1.55 eV with a repetition rate of 5 kHz at normal incidence [Fig. 1(a)]. Photons at this energy excite the metallic

Pb film to much higher excess energy density than the substrate because for Si the large direct bandgap of 3.4 eV and the absorption length of 12.7 μm ²⁹ suppress effective optical excitation of the substrate. The variation of the laser pulse energy by a factor of ≈ 25 resulted in a variation of the absorbed energy density Φ_{abs} in the Pb films from 6 to 140 J/cm^3 . Since the diameter of the pump laser beam (6 mm) was much larger than the width of the sample (2 mm), a homogeneous excitation of the film was ensured. The sample base temperature of the instrument was measured by a Si diode (Lake Shore Cryotronics DT-670C-SD) soldered by In to a different Si sample with the same size and specifications as used during the experiments. Upon liquid He cooling, we determined a minimum temperature of 19 K under high vacuum conditions for this sample. During all experiments, the Cu mount connected to the cold head of the cryostat was as cold as 9 K as determined by another Si diode (Lake Shore Cryotronics DT-670B-CU-HT). Due to heating by radiation and through the wires to the Si diodes, we expect the real sample base temperature to be below 19 K and above 9 K. Based on these constraints and the results of our data analysis, we estimated a sample base temperature of $T_{\text{Si}} \approx 10_{-1}^{+5}$ K. The difference in heat between 9 and 19 K for the electron and lattice system amounts to 3.8 J/cm^3 which is far below the applied absorbed energy densities in the experiment. The high thermal conductivity of up to ≈ 3500 $\text{W}/\text{m K}$ ^{30,31} of slightly n -doped Si at T_{Si} renders the substrate an almost perfect sink for the dissipated heat from the Pb film, i.e., the substrate's temperature rise was on the order of 1–2 K only.³² Further details on the experimental setup can be found elsewhere.^{16,33,34}

The Debye–Waller effect, $I = I_0 \exp\left(-\frac{1}{3} \langle \mathbf{u}^2 \rangle \cdot \Delta \mathbf{k}^2\right)$, which describes the intensity I of a diffraction spot with isotropic mean squared vibrational displacements $\langle \mathbf{u}^2 \rangle$ of the atoms and momentum transfer $\Delta \mathbf{k}$ is employed in URHEED at an electron energy of 30 keV to follow the transient temperature T_{Pb} of the Pb films upon optical excitation in a pump–probe setup as sketched in Fig. 1(a). We used a grazing angle of incidence of $\theta = 2.5^\circ \pm 0.1^\circ$ to provide surface sensitivity, i.e., $|\Delta \mathbf{k}| = 7.6 \pm 0.3$ \AA^{-1} for the (00) spot. T_{Pb} is determined from $\langle \mathbf{u}^2 \rangle$ in the framework of the Debye model. In order to account for the strong thermal lattice expansion of Pb³⁵ we used the temperature-dependent equivalent Debye temperature $\Theta_{\text{Pb},b}$ of bulk Pb³⁶ for the analysis. We also took into account that in the URHEED

experiment, mostly the topmost atomic layer of the Pb films was probed, which exhibits a reduced surface Debye temperature $\Theta_{\text{Pb},s} \approx 0.68\Theta_{\text{Pb},b}$,^{37,38} while the underlying layers were considered as bulk. Thus, the (00) spot intensity at temperature T_{Pb} is described by

$$\frac{I(T_{\text{Pb}})}{I_0} = \exp \left[-\gamma \left(1 + \frac{4T_{\text{Pb}}^2}{\Theta_{\text{Pb},s}^2} \int_0^{\Theta_{\text{Pb},s}} \frac{x dx}{e^x - 1} \right) \right], \quad (1)$$

where $\gamma = 3\hbar^2 \Delta \mathbf{k}^2 / 4m_{\text{Pb}} k_{\text{B}} \Theta_{\text{Pb},s}$ and m_{Pb} is the mass of Pb atoms.

The lattice excitation occurs within 3 ps subsequent to femtosecond optical irradiation through electron-phonon coupling of the photoexcited carriers followed by equilibration of the phonons through anharmonic coupling in 20–90 ps.³⁹ While the details of the excitation process are beyond the scope of this Letter, the thermal boundary conductance G is determined by the cooling behavior of the film. Thus, the evolution of film temperature T_{Pb} is evaluated through the transient intensity at time delay Δt between laser pump and electron probe pulse by fitting an exponential function for the recovery as

$$I(\Delta t)/I(T_{\text{Si}}) = \delta I_{\text{exc}}(\Delta t) e^{-\Delta t/\tau_{\text{rec}}}, \quad (2)$$

where δI is a fitting parameter for the maximum intensity drop, f_{exc} describes the excitation process of the lattice, and τ_{rec} is the recovery time constant.

As the substrate temperature remains almost constant, G can be determined by^{16,32,40}

$$G = -\frac{c_{\text{Pb}} \rho_{\text{Pb}} d}{T_{\text{Pb}} - T_{\text{Si}}} \frac{\partial T_{\text{Pb}}}{\partial \Delta t}, \quad (3)$$

where c_{Pb} is the specific heat capacity, ρ_{Pb} is the mass density, and d is the thickness of the Pb film. In the high-temperature limit $T_{\text{Pb}} \gg T_{\text{Si}}$, the solution of the differential equation for T_{Pb} results in an exponential decrease of the film temperature with a time constant $\tau_{\text{cool}} \approx \tau_{\text{rec}}$. Under these conditions, Eq. (3) simplifies to

$$G = \frac{c_{\text{Pb}} \rho_{\text{Pb}} d}{\tau_{\text{cool}}}. \quad (4)$$

Of course, due to the large temperature rise ΔT in our experiment, c_{Pb} , ρ_{Pb} , and d are not constant during the cooling of the film from T_{Pb} to

T_{Si} .^{35,41} However, for simplicity in our analysis we used the values at maximum film temperature $T_{\text{Si}} + \Delta T_{\text{max}}$. Furthermore, electronic heat conduction and electron-phonon scattering at the interface are considered negligible.

For comparison with theory, we use the semiclassical diffuse mismatch model (DMM).⁴ In DMM, phonons are considered to diffusely scatter at the interface conserving energy but not momentum or direction, allowing for arbitrary final momentum and direction [see inset of Fig. 3(a)]. The transmission probability Γ_{DMM} for phonons to cross the heterointerface is then given by Fermi's golden rule: the ratio of PDOS in film and substrate determines the final state in diffuse scattering, i.e., transmission toward the substrate or backscattering into the film. Taking into account the absence of optical phonon branches in Pb, the phonon transmission probability from the Pb film into the Si substrate is given in the Debye model (where $\text{PDOS}_i \propto v_i^{-3}$) by

$$\Gamma_{\text{DMM}} = \frac{1}{2} \left(1 + 2 \frac{v_{\text{Pb},l}^{-2} + 2v_{\text{Pb},t}^{-2}}{v_{\text{Si},l}^{-2} + 2v_{\text{Si},t}^{-2}} \right)^{-1}, \quad (5)$$

where v_i are the longitudinal (l) and transversal (t) sound velocities of the Pb film and the Si substrate along the [111] direction. We used bulk values for the phonon density of states and heat capacity as Monte Carlo simulations have shown that heterofilms consisting of only four MLs exhibit bulk-like properties.⁴²

Figure 1(b) depicts a RHEED pattern of a 6 ML Pb film. The transient normalized intensity $I(\Delta t)/I(T_{\text{Si}})$ of the (00) spot is shown in Fig. 2(a) for different absorbed energy densities Φ_{abs} , while a video of $\Delta I(\Delta t)/I(T_{\text{Si}})$ for the whole diffraction pattern is shown in the [supplementary material](#). Upon laser excitation at time delay $\Delta t = 0$, the intensity decreases on a timescale of 26–67 ps (time delay of minimum intensity) depending on the excitation strength. The recovery of the intensity to $I(T_{\text{Si}})$ (dashed gray line) occurs on a much slower timescale of several hundred picoseconds. The experimental data were fitted using Eq. (2) and the fits are shown as solid lines in Fig. 2(a). The observed intensity drop becomes larger with increasing Φ_{abs} , i.e., indicating an increase in film temperature T_{Pb} . The maximum rise of film temperature ΔT_{max} as determined from the maximum intensity drop $\Delta I_{\text{max}}/I(T_{\text{Si}})$ is indicated by short colored lines on the left ordinate in Fig. 2(a) and ranges from 10.6 K for the lowest to 94.8 K for the

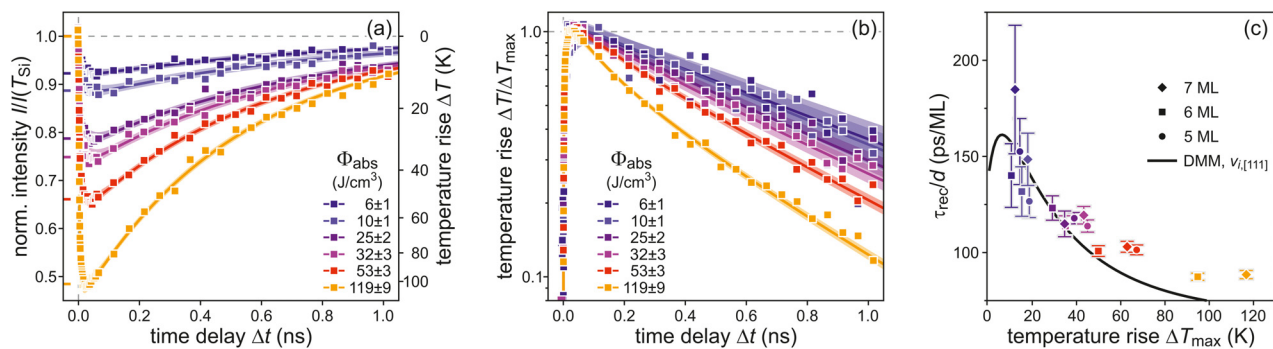


FIG. 2. Lattice dynamics of ultrathin Pb films: (a) Excitation and recovery of the normalized intensity $I(\Delta t)/I(T_{\text{Si}})$ of a 6 ML thin film using different absorbed energy densities Φ_{abs} . (b) Corresponding transient normalized temperature change $\Delta T(\Delta t)/\Delta T_{\text{max}}$. (c) Fraction of recovery time constant τ_{rec} and film thickness d as function of temperature rise ΔT_{max} from experiment (data points) and from DMM (black curve). The color of the symbols indicates the excitation strength, with blue corresponding to weak and orange to strong excitation. The shaded areas in panels (a) and (b) are the 1σ uncertainties of the fits.

highest absorbed energy density. In Fig. 2(b), the transient intensity $I(\Delta t)$ is converted into the normalized temperature change $\Delta T(\Delta t)/\Delta T_{\max}$. The fit of $I(\Delta t)/I(T_{\text{Si}})$ yields the time constants τ_{rec} corresponding to the recovery of transient intensity. As shown in Fig. 2(c), when τ_{rec} is normalized by the film thickness d , the data collapse onto a single curve, which is well described by the DMM (black line). The cooling significantly accelerates with increasing Φ_{abs} , i.e., τ_{rec} ranges from 840 ± 99 ps for the lowest to 524 ± 11 ps for the highest value of Φ_{abs} at $d = 6$ ML.

We determined G by employing Eq. (4) as shown in Fig. 3(a) for film thicknesses of 5–7 ML. The data for all three thicknesses collapse onto a single curve, which exhibits a clear dependence on the maximum film temperature $T_{\text{Pb,max}}$, i.e., on the absorbed energy density, with a steeper rise at lower ΔT and a significantly slower rise at higher ΔT . For $d = 6$ ML, the values range from $G = 1.29 \pm 0.18$ MW/m² K for the lowest to 4.02 ± 0.09 MW/m² K for the highest film temperature rise. The experimental values nicely match the theoretical expectation for the DMM for temperature increases up to $\Delta T \approx 50$ K. The

slight deviation between experiment and DMM at higher $T_{\text{Pb,max}}$ might arise from the simplicity of the applied model. More complex descriptions^{43–46} considering the real PDOS and dispersion of the phonons, as well as anharmonic contributions beyond the ones captured by the temperature dependencies of c_{Pb} , $\Theta_{\text{Pb,b}}$, q_{Pb} , d and v_i , and also inelastic phonon scattering might further improve the agreement.

Applying the DMM is justified because preparation of the Si(111)- $\beta(\sqrt{3} \times \sqrt{3})$ R30°-Pb wetting layer prior to film growth results in a surface alloy. During preparation at 600 °C, part of the Pb atoms desorbed, reducing the coverage from the ideal 1/3 ML to only 0.28 ML, as evident from the Si atoms in substitutional sites seen in the scanning tunneling microscopy (STM) image in Fig. 3(b). The accompanying loss of long-range order and increased local disorder are reflected in the spot profile analysis low-energy electron diffraction (SPA-LEED) pattern shown in Fig. 3(c), which exhibits diffuse honeycomb intensity at (3×3) positions. This surface alloy of heavy Pb and light Si atoms induces diffuse scattering, relaxing momentum conservation of phonons at the interface and hereby necessitating the application of DMM.

The role of crystallinity, grain size, disorder, and defects in the Pb film, as well as the sharpness and well-defined nature of the interface on the thermal boundary conductance G could be assessed by comparison with Pb films grown under different experimental conditions. Lyeo and Cahill¹ studied Pb films grown at room temperature on a H-terminated Si(111) substrate after HF treatment. Such a preparation results in strongly textured Pb(111) films due to a much rougher Si surface.^{47,48} H termination results in a significantly more complex interface with modified properties, which leads to a higher phonon transmission probability. The resulting value for G by Lyeo and Cahill¹ of ≈ 14 MW/m² K at 90–120 K is more than three times higher than in our measurements on monocrystalline epitaxial Pb films with a well-defined alloyed but sharp interface. Thus, high-quality substrate and film morphologies are crucial for lowering the thermal boundary conductance of the Pb/Si interface to the ultimate limit.

In summary, the transient heating of ultrathin epitaxial Pb films on a Si(111) substrate was studied utilizing femtosecond laser pulses for pumping and picosecond electron pulses for probing. The temperature evolution was analyzed by applying the Debye–Waller effect to the measured decline in URHEED intensity. From the data, we derive the thermal boundary conductance G of the interface at the maximum film temperature, specifically at large temperature discontinuities up to $\Delta T \approx 120$ K. The temperature dependence of the thermal boundary conductance aligns well with predictions from the simple diffuse mismatch model up to moderate excitation densities of $\Phi_{\text{abs}} \approx 50$ J/cm². The thermal boundary conductance of our high-quality epitaxial Pb films grown on an atomically perfectly ordered Si substrate is much lower than that of films grown at room temperature on a rough Si surface, indicating the crucial role of substrate, film, and interface morphologies on heat transfer.

See the [supplementary material](#) for a video illustrating the transient intensity change in URHEED of a 6 ML Pb film at the highest absorbed energy density.

We acknowledge the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) for financial support via projects B04 and C03 of the Collaborative Research Center SFB1242

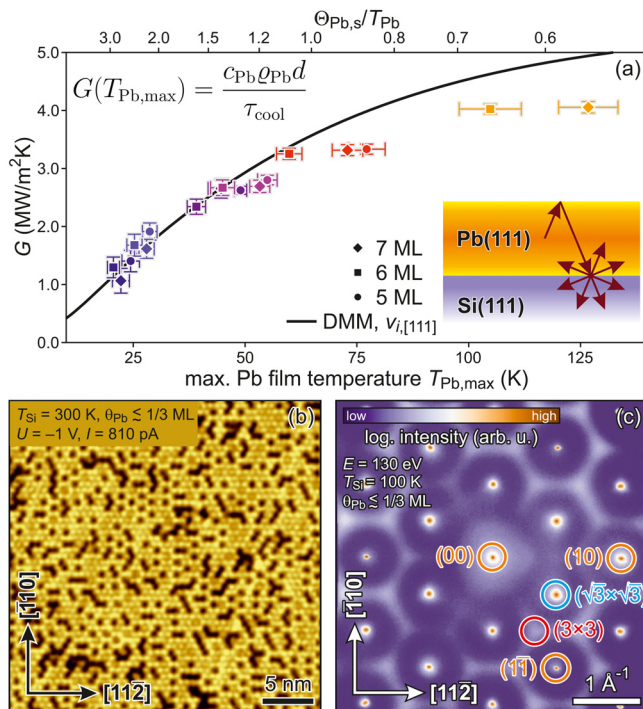


FIG. 3. Thermal boundary conductance G and structure of the wetting layer: (a) Experimental G for film thicknesses of 5–7 ML, compared to the DMM in Debye model with the sound velocities along the [111] direction (black curve). The color of the symbols indicates the excitation strength, with blue corresponding to weak and orange to strong excitation. Inset: Scattering mechanism for phonons in DMM. (b) STM image of the Si(111)- $\beta(\sqrt{3} \times \sqrt{3})$ R30°-Pb wetting layer at room temperature taken at negative bias of -1 V with a current of 810 pA, i.e., probing occupied states. Bright features correspond to Pb atoms while dark features indicate Si atoms in substitutional sites replacing the Pb atoms. The Pb coverage is reduced to 0.28 ML instead of 1/3 ML. (c) SPA-LEED pattern of the Si(111)- $\beta(\sqrt{3} \times \sqrt{3})$ R30°-Pb wetting layer at 100 K taken at 130 eV. The formation of the Pb/Si surface alloy and increased disorder causes the honeycomb-shaped diffuse intensity at (3×3) positions.

“Nonequilibrium dynamics of condensed matter in the time domain” (Project-ID 278162697) and via the project “Transport and collective excitations in metallic wires” of the Research Unit FOR1700 “Metallic nanowires on the atomic scale: Electronic and vibrational coupling in real world systems” (Project-ID 194370842). The authors thank D. G. Cahill, B. Rethfeld, and Th. Groven for stimulating discussions.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Christian Brand: Conceptualization (equal); Data curation (lead); Formal analysis (lead); Investigation (equal); Visualization (lead); Writing – original draft (lead); Writing – review & editing (lead). **Tobias Witte:** Formal analysis (equal); Investigation (equal); Writing – original draft (supporting); Writing – review & editing (supporting). **Mohammad Tajik:** Formal analysis (supporting); Writing – original draft (supporting); Writing – review & editing (supporting). **Jonas D. Fortmann:** Writing – original draft (supporting); Writing – review & editing (supporting). **Birk Finke:** Writing – original draft (supporting); Writing – review & editing (supporting). **Herbert Pfnür:** Funding acquisition (supporting); Project administration (supporting); Supervision (supporting). **Christoph Tegenkamp:** Funding acquisition (supporting); Project administration (supporting); Supervision (supporting). **Michael Horn-von Hoegen:** Conceptualization (lead); Funding acquisition (lead); Methodology (lead); Project administration (lead); Supervision (lead); Visualization (supporting); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

REFERENCES

- H.-K. Lyeo and D. G. Cahill, “Thermal conductance of interfaces between highly dissimilar materials,” *Phys. Rev. B* **73**, 144301 (2006).
- D. G. Cahill, W. K. Ford, K. E. Goodson, G. D. Mahan, A. Majumdar, H. J. Maris, R. Merlin, and S. R. Phillpot, “Nanoscale thermal transport,” *J. Appl. Phys.* **93**, 793–818 (2003).
- D. G. Cahill, P. V. Braun, G. Chen, D. R. Clarke, S. Fan, K. E. Goodson, P. Keblinski, W. P. King, G. D. Mahan, A. Majumdar *et al.*, “Nanoscale thermal transport. II. 2003–2012,” *Appl. Phys. Rev.* **1**, 011305 (2014).
- E. T. Swartz and R. O. Pohl, “Thermal boundary resistance,” *Rev. Mod. Phys.* **61**, 605–668 (1989).
- D. G. Cahill, “Thermal conductivity measurement from 30 to 750 K: The 3 ω method,” *Rev. Sci. Instrum.* **61**, 802–808 (1990).
- D. G. Cahill, “Analysis of heat flow in layered structures for time-domain thermoreflectance,” *Rev. Sci. Instrum.* **75**, 5119–5122 (2004).
- A. J. Schmidt, “Pump-probe thermoreflectance,” *Annu. Rev. Heat Transfer* **16**, 159–181 (2013).
- A. J. Schmidt, X. Chen, and G. Chen, “Pulse accumulation, radial heat conduction, and anisotropic thermal conductivity in pump-probe transient thermoreflectance,” *Rev. Sci. Instrum.* **79**, 114902 (2008).
- M. Chang, Z. Yuan, N. Liang, X. Fan, Y. Wang, J. Li, Y. Deng, X.-J. Yan, M.-H. Lu, B. Song, and H. Lu, “Interfacial thermal transport suppression by hydrogen insertion in an epitaxial Al/Si heterostructure,” *Appl. Phys. Lett.* **126**, 212201 (2025).
- J. A. Malen, K. Baheti, T. Tong, Y. Zhao, J. A. Hudgings, and A. Majumdar, “Optical measurement of thermal conductivity using fiber aligned frequency domain thermoreflectance,” *J. Heat Transfer* **133**, 081601 (2011).
- K. T. Regner, S. Majumdar, and J. A. Malen, “Instrumentation of broadband frequency domain thermoreflectance for measuring thermal conductivity accumulation functions,” *Rev. Sci. Instrum.* **84**, 064901 (2013).
- J. Yang, C. Maragliano, and A. J. Schmidt, “Thermal property microscopy with frequency domain thermoreflectance,” *Rev. Sci. Instrum.* **84**, 104904 (2013).
- D. G. Cahill and R. O. Pohl, “Thermal conductivity of amorphous solids above the plateau,” *Phys. Rev. B* **35**, 4067–4073 (1987).
- C. Dames, “Measuring the thermal conductivity of thin films: 3 ω and related electrothermal methods,” *Annu. Rev. Heat Transfer* **16**, 7–49 (2013).
- M. Hu, P. Keblinski, J.-S. Wang, and N. Ravivkar, “Interfacial thermal conductance between silicon and a vertical carbon nanotube,” *J. Appl. Phys.* **104**, 083503 (2008).
- B. Krenzer, A. Janzen, P. Zhou, D. von der Linde, and M. Horn-von Hoegen, “Thermal boundary conductance in heterostructures studied by ultrafast electron diffraction,” *New J. Phys.* **8**, 190 (2006).
- A. Hanisch, B. Krenzer, T. Pelka, S. Möllenbeck, and M. Horn-von Hoegen, “Thermal response of epitaxial thin Bi films on Si(001) upon femtosecond laser excitation studied by ultrafast electron diffraction,” *Phys. Rev. B* **77**, 125410 (2008).
- A. Hanisch-Blicharski, A. Janzen, B. Krenzer, S. Wall, F. Klasing, A. Kalus, T. Frigge, M. Kammler, and M. Horn-von Hoegen, “Ultra-fast electron diffraction at surfaces: From nanoscale heat transport to driven phase transitions,” *Ultramicroscopy* **127**, 2–8 (2013).
- T. Frigge, B. Hafke, V. Tinnemann, B. Krenzer, and M. Horn-von Hoegen, “Nanoscale heat transport from Ge hut, dome, and relaxed clusters on Si(001) measured by ultrafast electron diffraction,” *Appl. Phys. Lett.* **106**, 053108 (2015).
- T. Witte, T. Frigge, B. Hafke, B. Krenzer, and M. Horn-von Hoegen, “Nanoscale interfacial heat transport of ultrathin epitaxial hetero films: Few monolayer Pb(111) on Si(111),” *Appl. Phys. Lett.* **110**, 243103 (2017).
- M. Gorfien, H. Wang, L. Chen, H. Rahmani, J. Yu, P. Zhu, J. Chen, X. Wang, J. Zhao, and J. Cao, “Nanoscale thermal transport across an GaAs/AlGaAs heterostructure interface,” *Struct. Dyn.* **7**, 025101 (2020).
- W. Braun, *Applied RHEED: Reflection High-Energy Electron Diffraction During Crystal Growth* (Springer Science & Business Media, 1999), Vol. 154.
- P. Debye, “Interferenz von Röntgenstrahlen und Wärmebewegung,” *Ann. Phys.* **348**, 49–92 (1913).
- I. Waller, “Zur Frage der Einwirkung der Wärmebewegung auf die Interferenz von Röntgenstrahlen,” *Z. Phys.* **17**, 398–408 (1923).
- P. Kury, R. Hild, D. Thien, H.-L. Günter, F.-J. Meyer zu Heringdorf, and M. Horn-von Hoegen, “Compact and transferable threefold evaporator for molecular beam epitaxy in ultrahigh vacuum,” *Rev. Sci. Instrum.* **76**, 083906 (2005).
- A. Petkova, J. Wollschläger, H.-L. Günter, and M. Henzler, “Order and disorder in ultrathin Pb films grown on Si(111) 7×7 substrates at low temperatures,” *Surf. Sci.* **482**, 922–927 (2001).
- A. Ichimiya and P. I. Cohen, “RHEED intensity oscillations,” in *Reflection High-Energy Electron Diffraction* (Cambridge University Press, 2004), pp. 270–313.
- M. Horn-von Hoegen, “Growth of semiconductor layers studied by spot profile analysing low energy electron diffraction – Part I,” *Z. Kristallogr.* **214**, 684–721 (1999).
- D. E. Aspnes and A. A. Studna, “Dielectric functions and optical parameters of Si, Ge, GaP, GaAs, GaSb, InP, InAs, and InSb from 1.5 to 6.0 eV,” *Phys. Rev. B* **27**, 985–1009 (1983).
- J. C. Thompson and B. A. Younglove, “Thermal conductivity of silicon at low temperatures,” *J. Phys. Chem. Solids* **20**, 146–149 (1961).
- C. J. Glassbrenner and G. A. Slack, “Thermal conductivity of silicon and germanium from 3 K to the melting point,” *Phys. Rev.* **134**, A1058–A1069 (1964).
- A. Hanisch-Blicharski, V. Tinnemann, S. Wall, F. Thiemann, T. Groven, J. Fortmann, M. Tajik, C. Brand, B.-O. Frost, A. von Hoegen, and M. Horn-von Hoegen, “Violation of Boltzmann equipartition theorem in angular phonon phase space slows down nanoscale heat transfer in ultrathin heterofilms,” *Nano Lett.* **21**, 7145–7151 (2021).

- ³³B. Hafke, T. Witte, C. Brand, Th. Duden, and M. Horn-von Hoegen, "Pulsed electron gun for electron diffraction at surfaces with femtosecond temporal resolution and high coherence length," *Rev. Sci. Instrum.* **90**, 045119 (2019).
- ³⁴M. Horn-von Hoegen, "Structural dynamics at surfaces by ultrafast reflection high-energy electron diffraction," *Struct. Dyn.* **11**, 021301 (2024).
- ³⁵T. Rubin, H. L. Johnston, and H. W. Altman, "The thermal expansion of lead," *J. Phys. Chem.* **66**, 266–268 (1962).
- ³⁶O. P. Gupta, "Thermal properties of lead," *Physica B+C* **122**, 236–240 (1983).
- ³⁷H. E. Elsayed-Ali, "Surface Debye temperature measurement with reflection high-energy electron diffraction," *J. Appl. Phys.* **79**, 6853–6857 (1996).
- ³⁸V. Tinnemann, C. Streubühr, B. Hafke, T. Witte, A. Kalus, A. Hanisch-Blicharski, M. Ligges, P. Zhou, D. von der Linde, U. Bovensiepen, and M. Horn-von Hoegen, "Decelerated lattice excitation and absence of bulk phonon modes at surfaces: Ultra-fast electron diffraction from Bi(111) surface upon fs-laser excitation," *Struct. Dyn.* **6**, 065101 (2019).
- ³⁹M. Tajik, T. Witte, C. Brand, L. Rettig, B. Sothmann, U. Bovensiepen, and M. Horn-von Hoegen, "Electron phonon coupling in ultrathin Pb films on Si(111): Where the heck is the energy?," [arXiv:2312.04541](https://arxiv.org/abs/2312.04541) (2023).
- ⁴⁰R. J. Stoner and H. J. Maris, "Kapitza conductance and heat flow between solids at temperatures from 50 to 300 K," *Phys. Rev. B* **48**, 16373–16387 (1993).
- ⁴¹P. F. Meads, W. R. Forsythe, and W. F. Giauque, "The heat capacities and entropies of silver and lead from 15° to 300 °K," *J. Am. Chem. Soc.* **63**, 1902–1905 (1941).
- ⁴²B. Krenzer, A. Hanisch-Blicharski, P. Schneider, Th. Payer, S. Möllenbeck, O. Osmani, M. Kammler, R. Meyer, and M. Horn-von Hoegen, "Phonon confinement effects in ultrathin epitaxial bismuth films on silicon studied by time-resolved electron diffraction," *Phys. Rev. B* **80**, 024307 (2009).
- ⁴³J. G. Chung, A. J. H. McGaughey, and M. Kaviani, "Role of phonon dispersion in lattice thermal conductivity modeling," *J. Heat Transfer* **126**, 376–380 (2004).
- ⁴⁴P. E. Hopkins, J. C. Duda, and P. M. Norris, "Anharmonic phonon interactions at interfaces and contributions to thermal boundary conductance," *J. Heat Transfer* **133**, 062401 (2011).
- ⁴⁵P. Reddy, K. Castelino, and A. Majumdar, "Diffuse mismatch model of thermal boundary conductance using exact phonon dispersion," *Appl. Phys. Lett.* **87**, 211908 (2005).
- ⁴⁶J. C. Duda, T. E. Beechem, J. L. Smoyer, P. M. Norris, and P. E. Hopkins, "Role of dispersion on phononic thermal boundary conductance," *J. Appl. Phys.* **108**, 073515 (2010).
- ⁴⁷H. Angermann, J. Rappich, L. Korte, I. Sieber, E. Conrad, M. Schmidt, K. Hübener, J. Polte, and J. Hauschild, "Wet-chemical passivation of atomically flat and structured silicon substrates for solar cell application," *Appl. Surf. Sci.* **254**, 3615–3625 (2008).
- ⁴⁸W. Henrion, M. Rebien, H. Angermann, and A. Röseler, "Spectroscopic investigations of hydrogen termination, oxide coverage, roughness, and surface state density of silicon during native oxidation in air," *Appl. Surf. Sci.* **202**, 199–205 (2002).