MRS Spring meeting San Francisco April 2014

Symposium A. Film-silicon science and technology

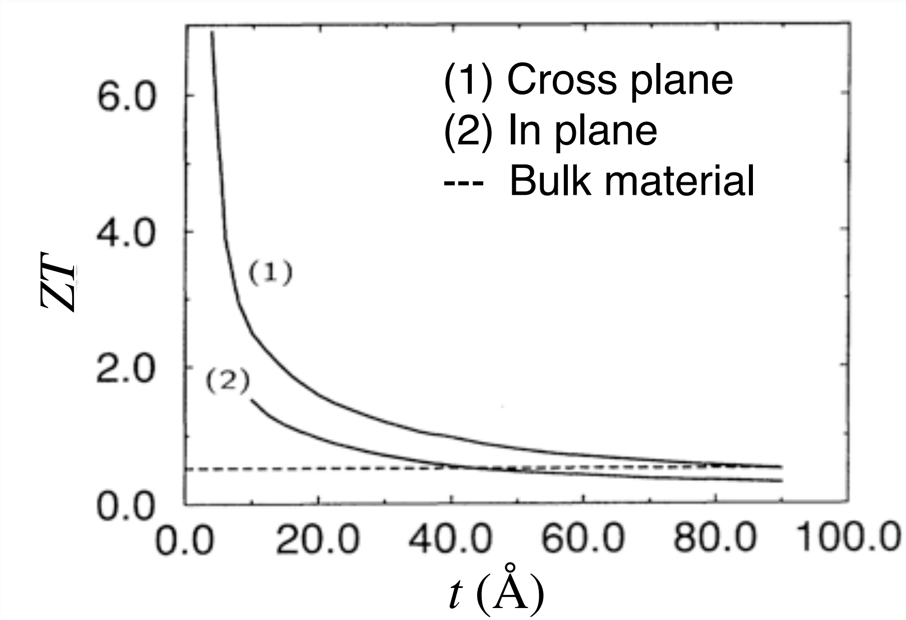
Silicon-based thin films and 0–3 composites with very low thermal conductivity

Hartmut S. Leipner, Peter Werner, Katrin Bertram, Markus Trutschel, Bodo Fuhrmann, Alexander Tonkikh

Martin-Luther-Universität Halle–Wittenberg

Max-Planck-Institut für Mikrostrukturphysik Halle

Control of thermoelectric properties

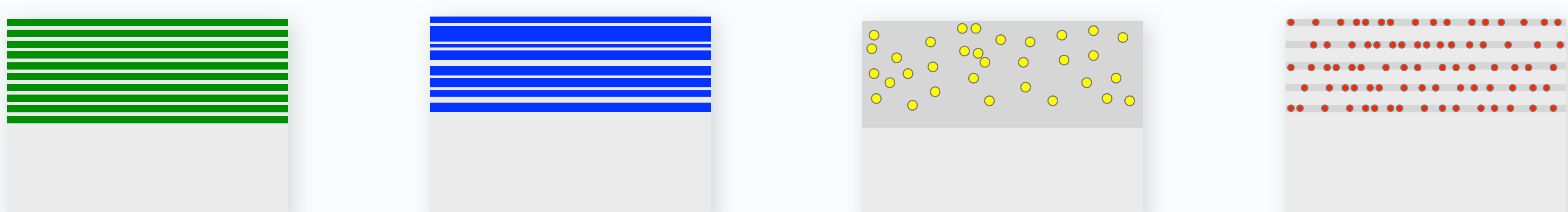


$$ZT = \frac{S^2\sigma}{\kappa}T$$

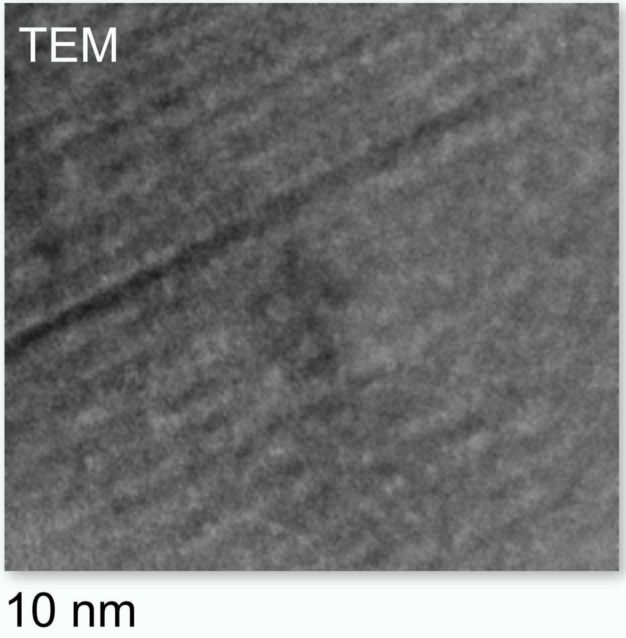
Thermoelectric figure of merit ZT for anisotropic Bi_2Te_3 layers of the thickness t

[Hicks, Dresselhaus 1993]

Superlattices, random multilayers, composites, quantum dot SLs

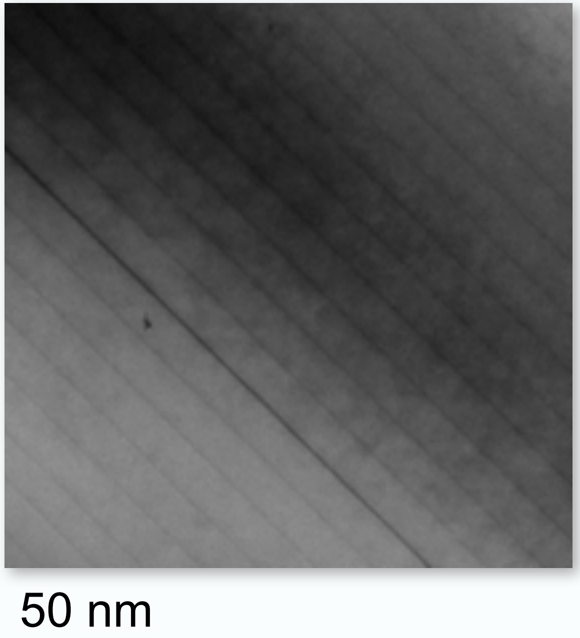


Si–Si$_{1-x}$Ge$_x$ superlattices



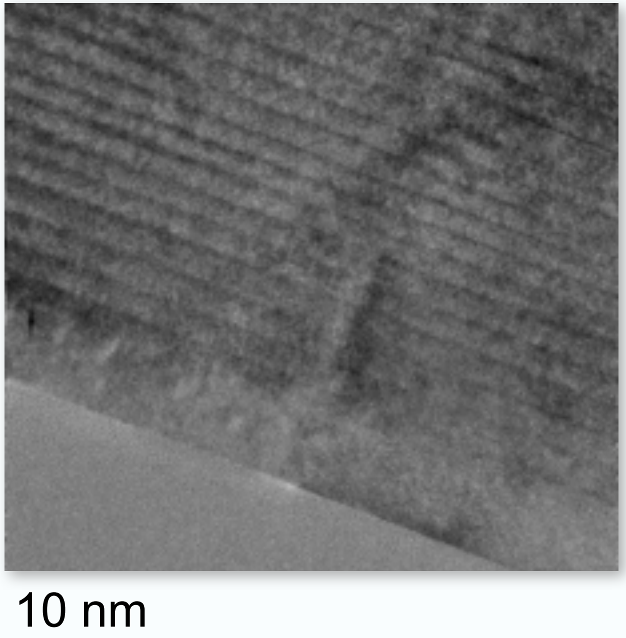
0.2 nm Ge + 3.3 nm Si

171×, ≈ 600 nm



1.6 nm Ge + 12 nm Si

39×, ≈ 600 nm



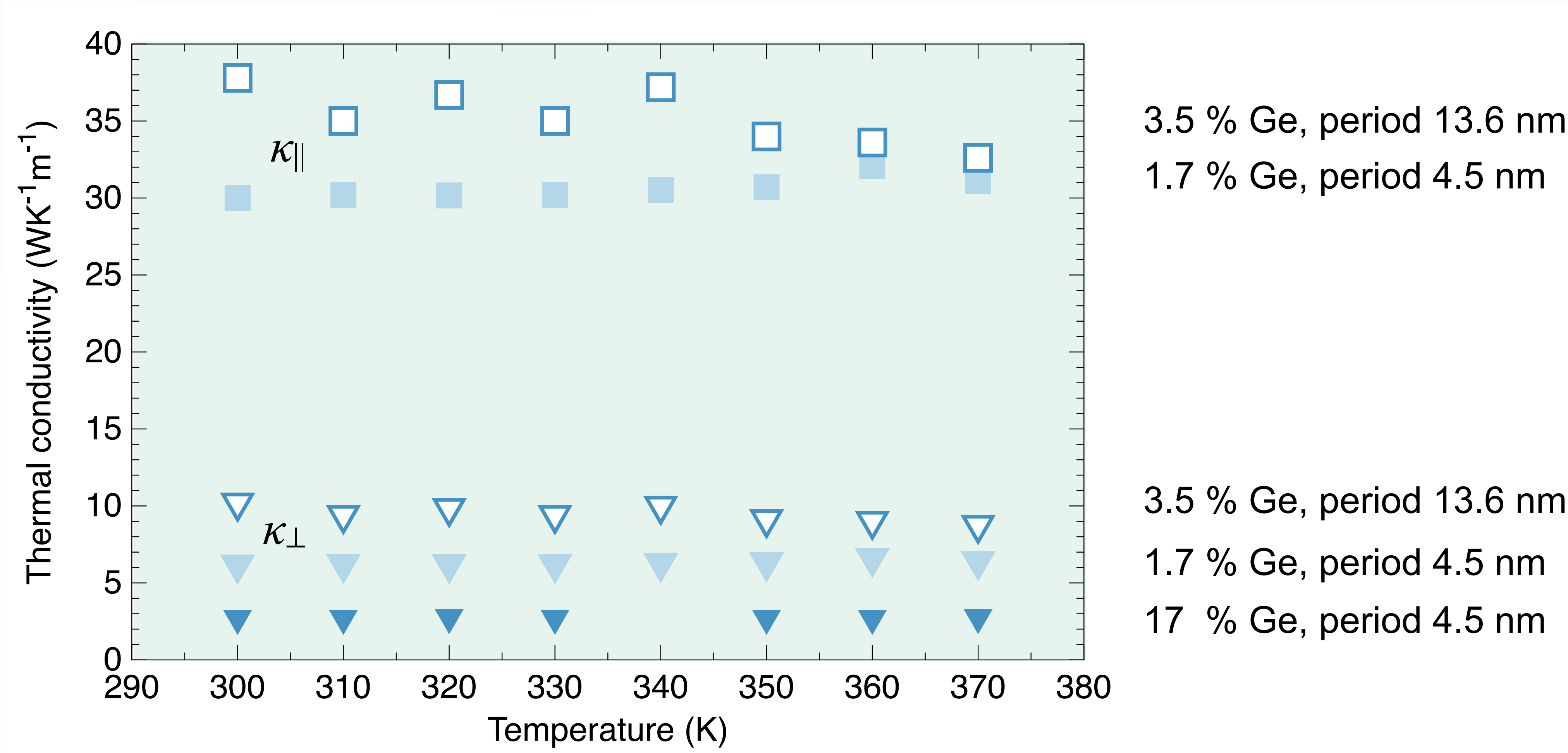
2 nm Ge + 1.5 nm Si

171×, ≈ 600 nm

Average Ge content

1.7 % 3.5 % 17 %

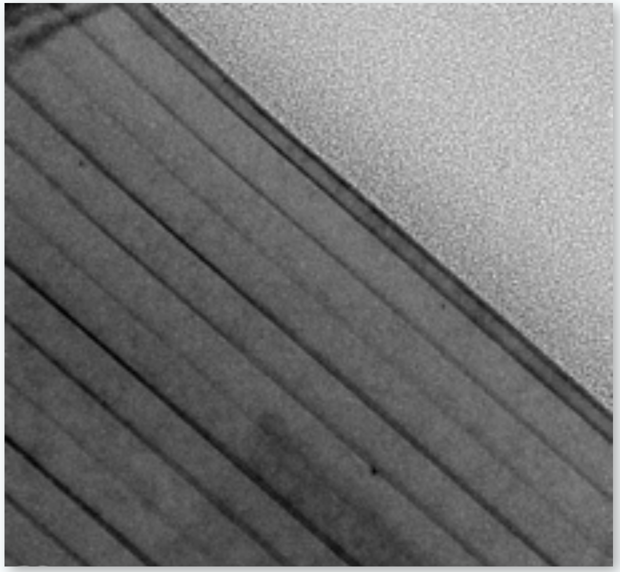
Thermal conductivity of superlattices

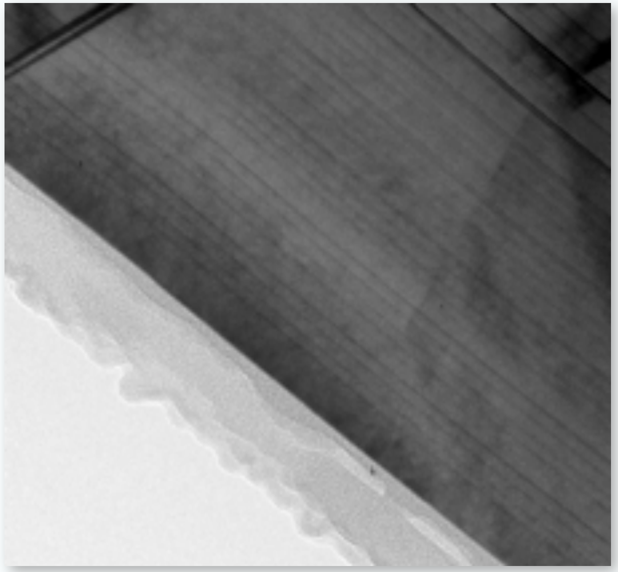


In-plane ($\kappa_{||}$) and cross-plane ($\kappa_{\perp}$) thermal conductivities for superlattices with different Ge contents and periods

Random multilayers

20 nm



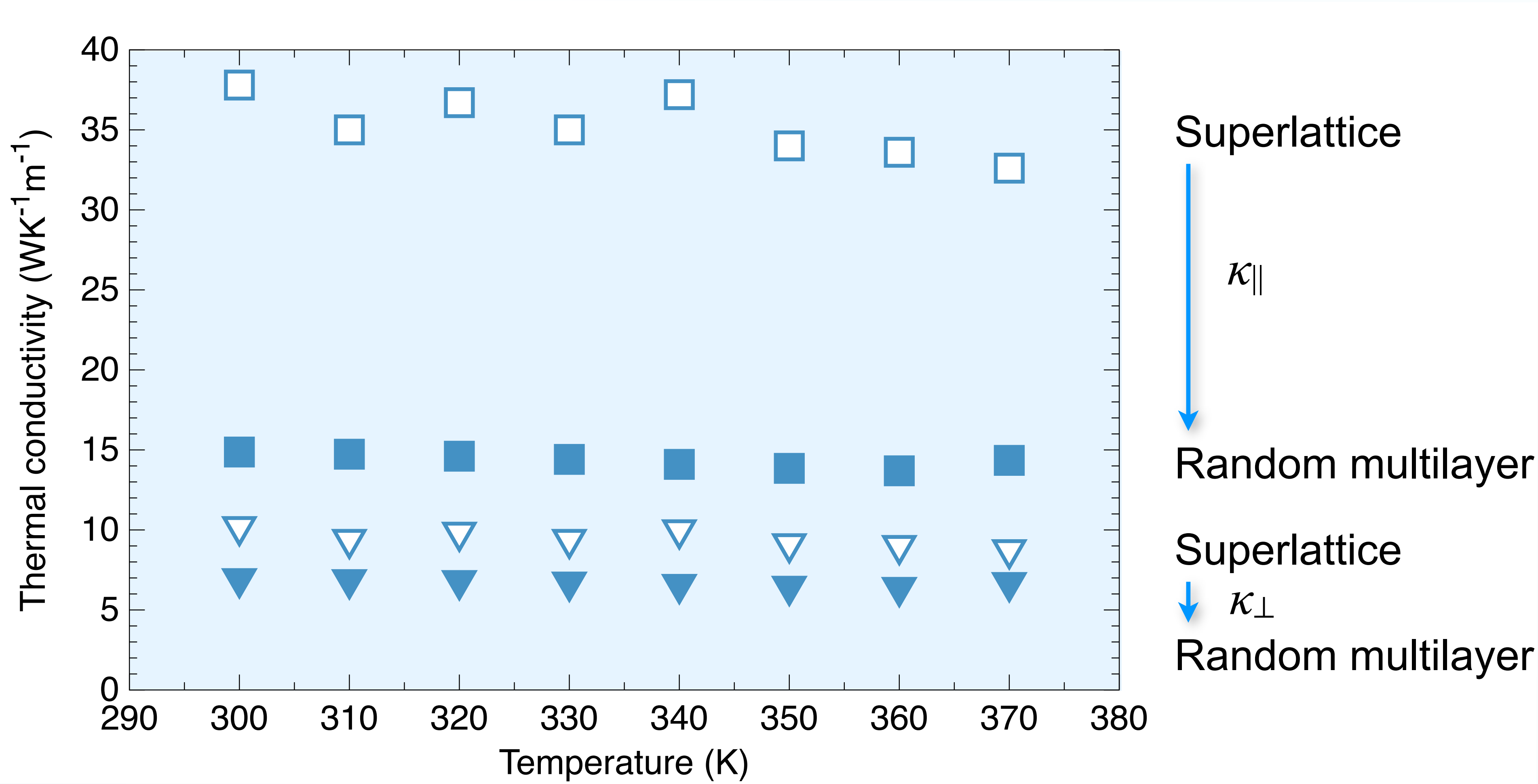


1.2 nm Ge + 12 nm Si	0.6 nm Ge + 4.1 nm Si
1.2 nm Ge + 12 nm Si	0.3 nm Ge + 5.1 nm Si
1.8 nm Ge + 12 nm Si	0.8 nm Ge + 4.8 nm Si
0.9 nm Ge + 12 nm Si	0.6 nm Ge + 5.7 nm Si
1.6 nm Ge + 12 nm Si	0.6 nm Ge + 3.8 nm Si
6×, ≈ 600 nm	34×, ≈ 940 nm

Average Ge content

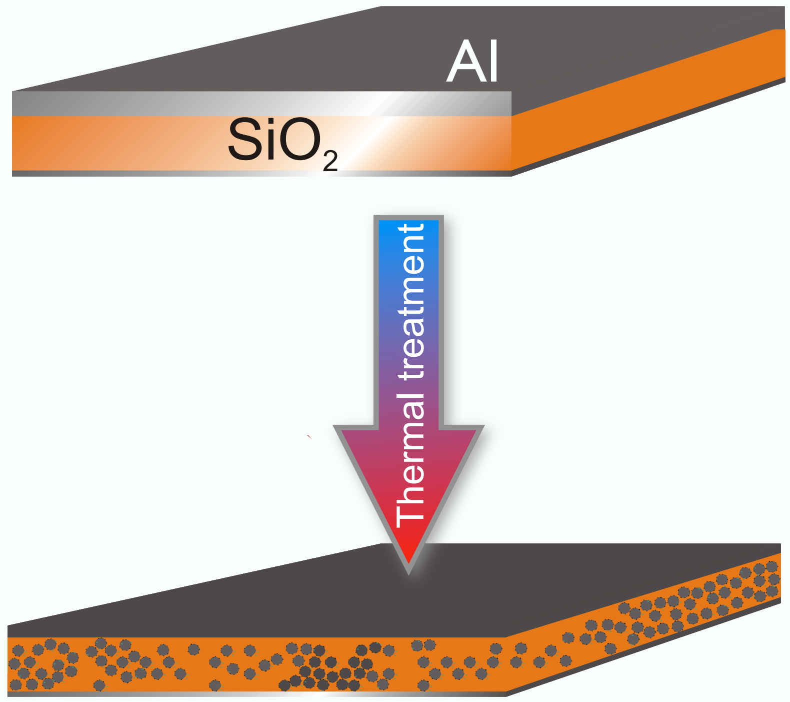
2.9 %	3.3 %

3ω results of random multilayers



Thermal conductivities in a random multilayer (2.9 % Ge)
in comparison to a superlattice (3.5 % Ge)

Nanoparticles in thin-film oxide

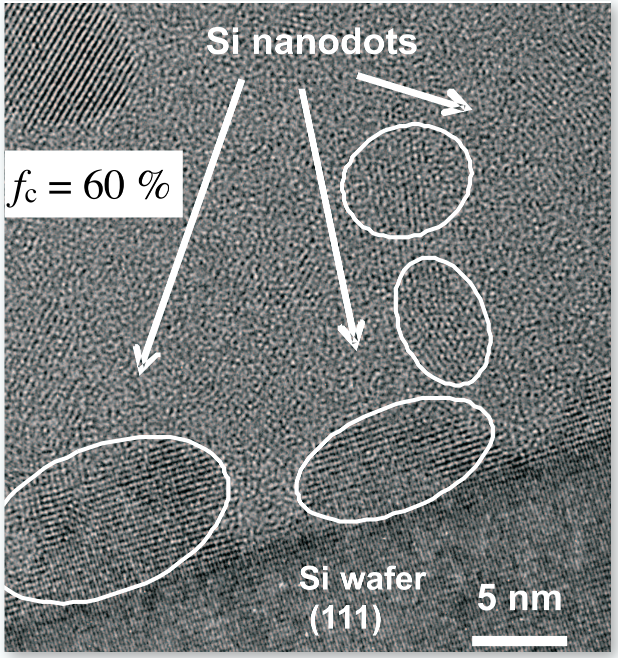


Al_2O_3 film with Si nanoparticles

Fabrication of thin film 0–3 composites

- Solid-state reaction $3\ SiO_2 + 4\ Al \rightarrow 3\ Si + 2\ Al_2O_3$
- PECVD deposition of SiO_x and subsequent crystallization to form Si nanodots at the percolation limit; $2\ SiO_x \rightarrow (2 - x)\ Si + x\ SiO_2$

Oxide-embedded Si nanodots



$SiO_{1.3}$

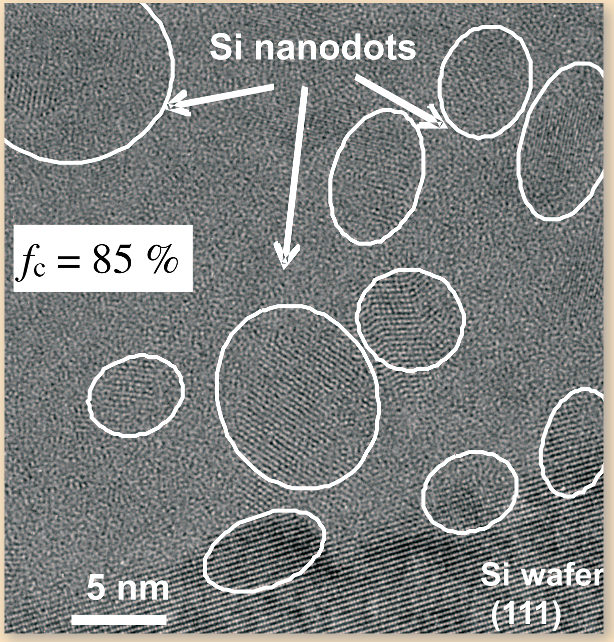
The degree of crystallization f_c depends on the oxygen content x in the SiO_x film.

[Roczen *et al* J Non-Cryst Sol 2011]

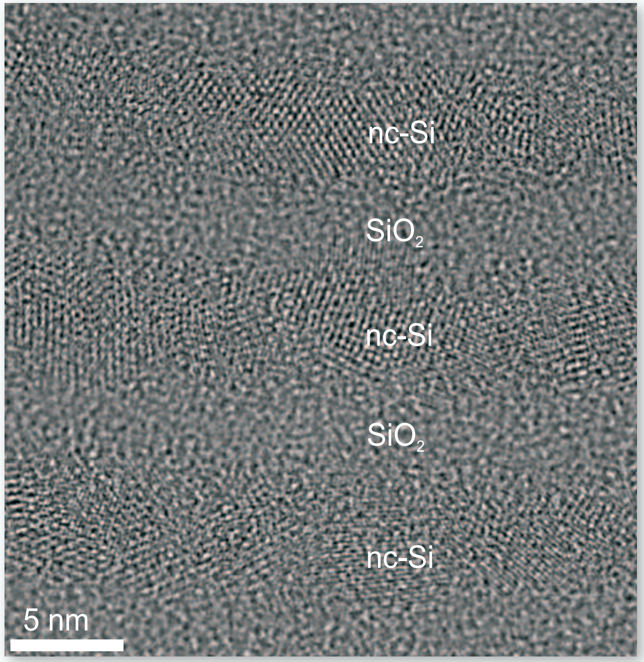
$$2\ SiO_x \rightarrow (2 - x)\ Si + x\ SiO_2$$

Layer structure of nc-Si in SiO_2

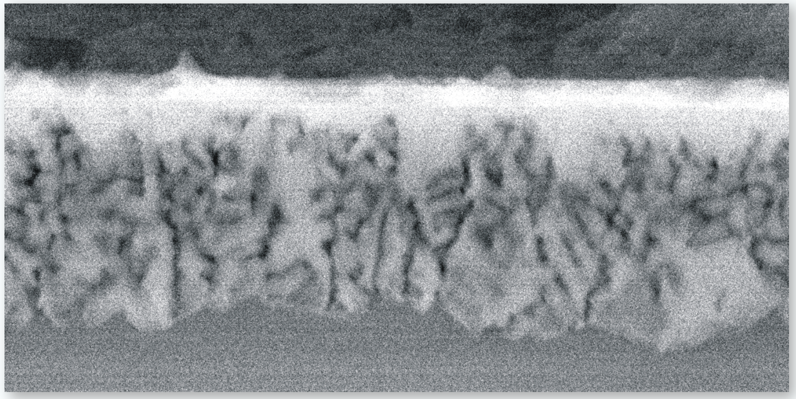
Percolation limit

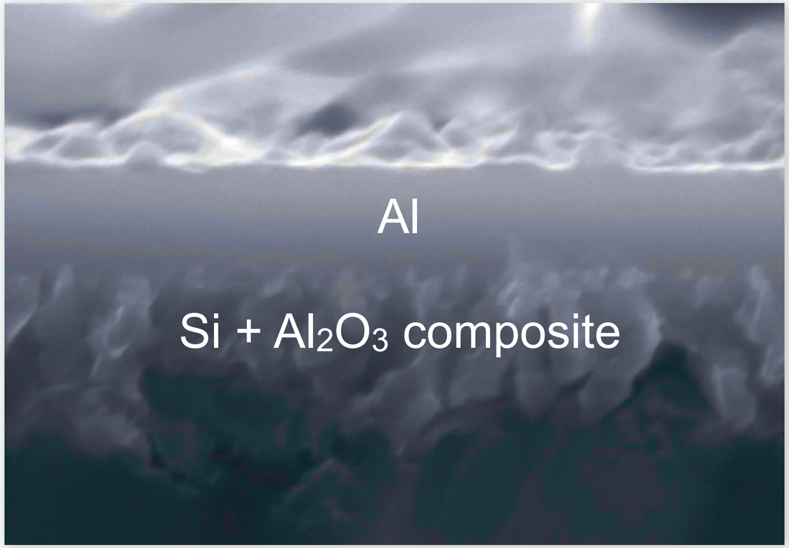


$SiO_{0.1}$



Synthesis of Si particles in Al_2O_3

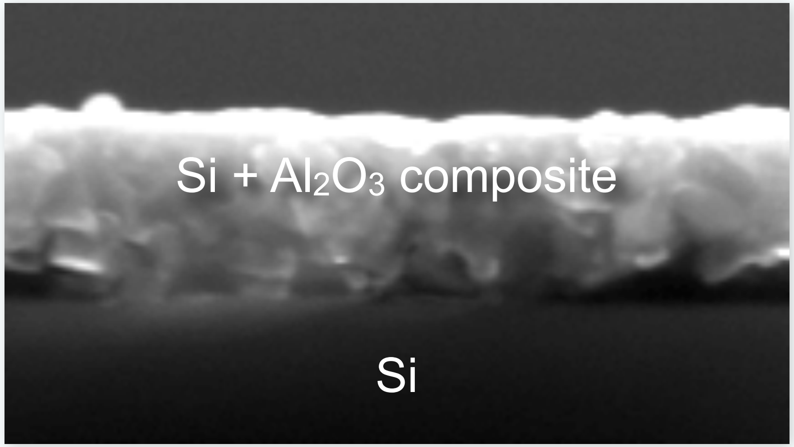




500 nm

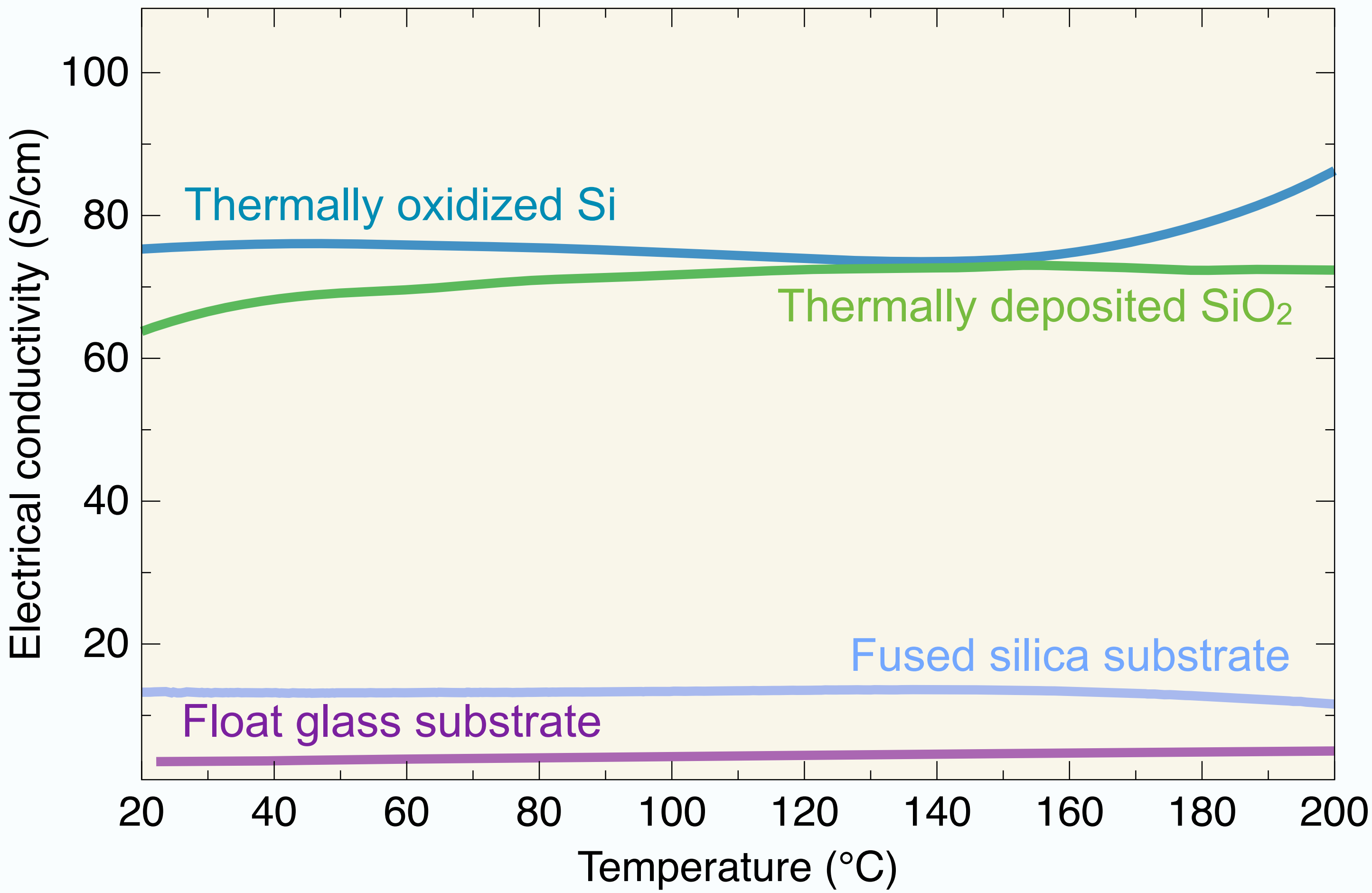
Process parameters

- Initial thicknesses d_{Al}, $d_{\mathrm{SiO_2}}$, temperature (500…600 °C), annealing time (1…3 h)
- Reaction rate ≈ 3 nm/min at 550 °C
- Different substrates



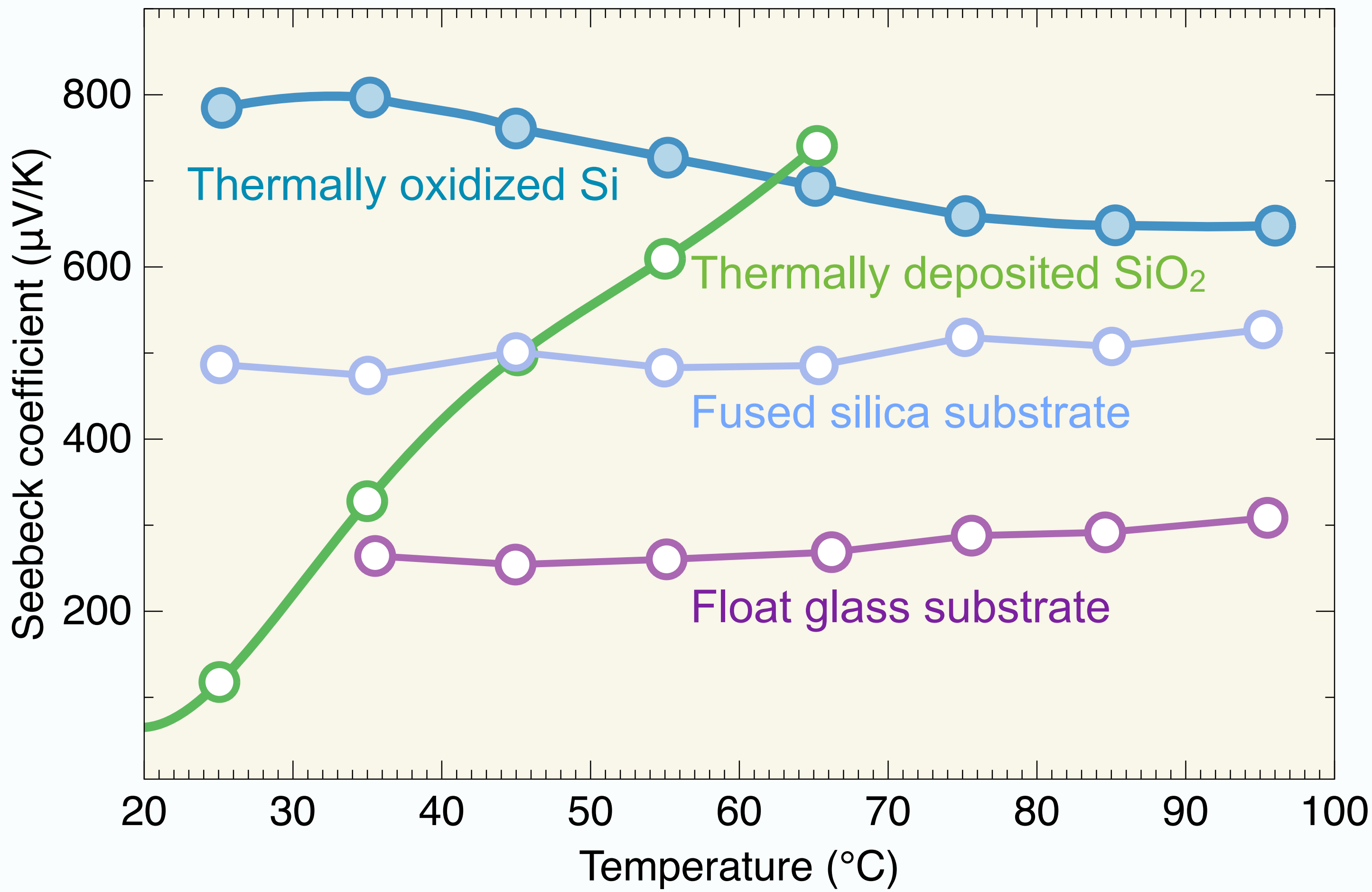
Electrical conductivity of Si–Al_2O_3 films

$$3\ SiO_2 + 4\ Al \rightarrow 3\ Si + 2\ Al_2O_3$$



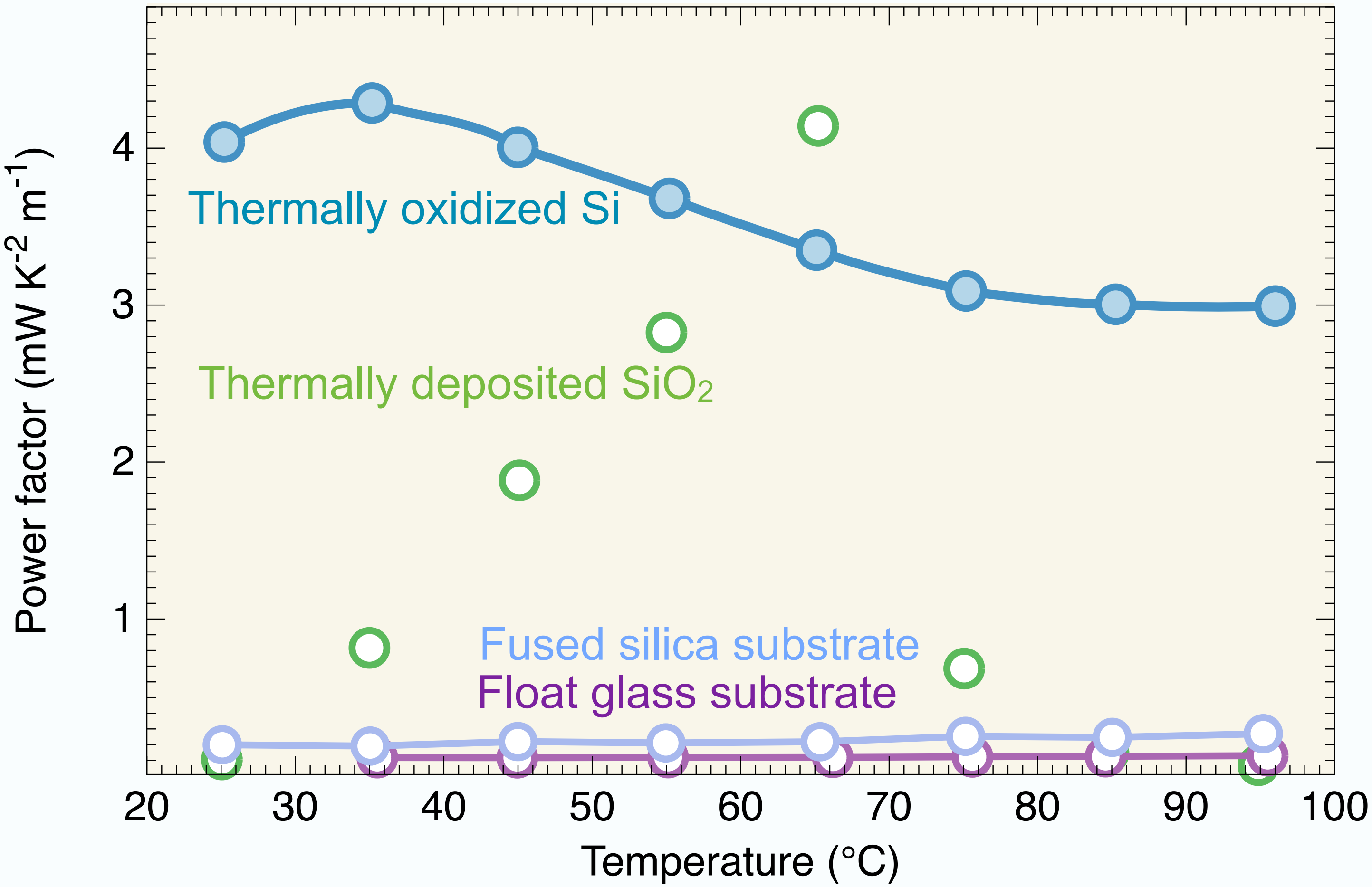
Electrical conductivity σ of the composite film for different substrates used

Seebeck measurements of $Si–Al_2O_3$ films



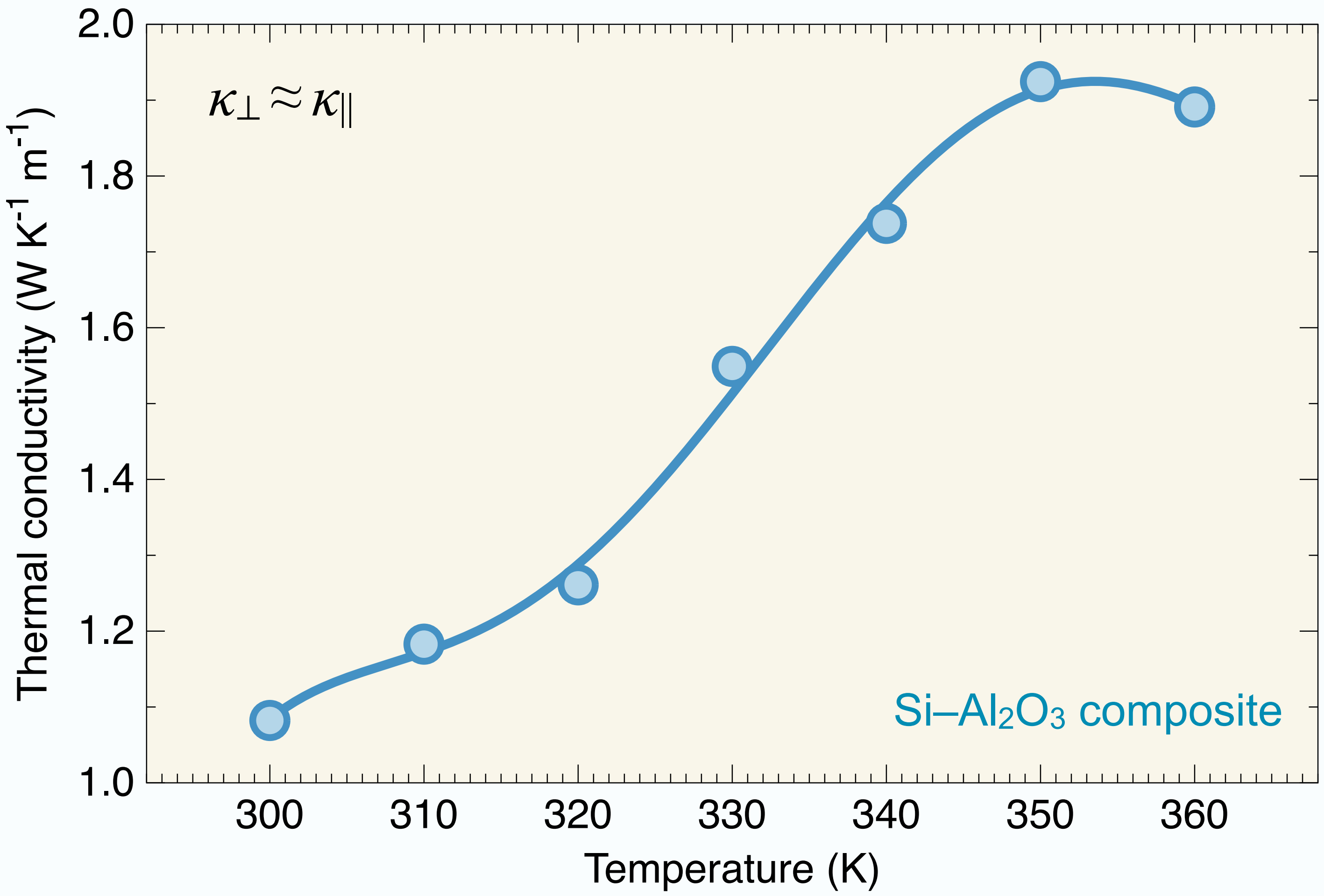
Seebeck coefficient *S* of the composite film for different substrates used

Power factor of $Si–Al_2O_3$ films



Power factor $S^2\sigma$ of the composite films

Comparison of thermal conductivities



Thermal conductivity κ of the composite film formed in thermally oxidized silicon

Comparison of $Si–Al_2O_3$ films

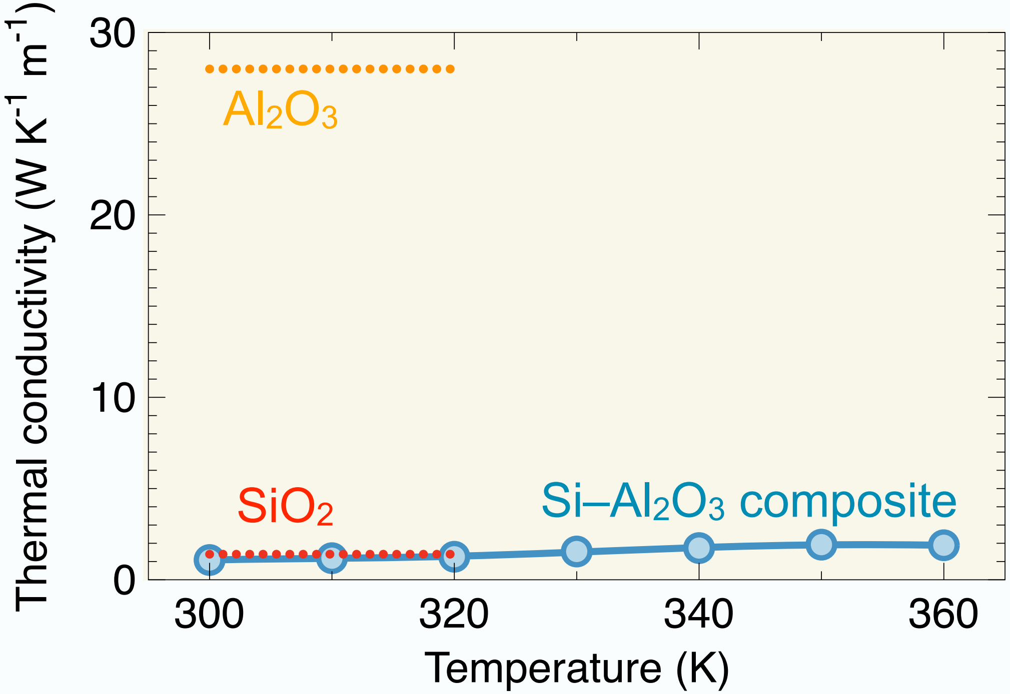
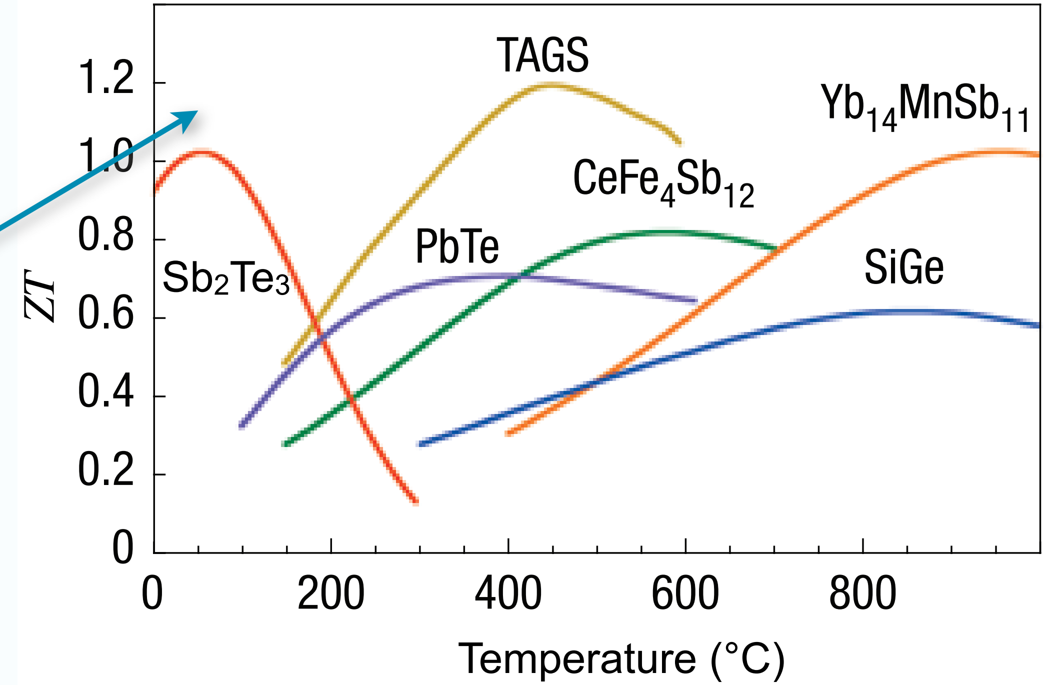


Figure of merit for different materials

[Snyder 2008]



ZT = 1.1 @ 300 K for $Si–Al_2O_3$ composites in thermally oxidized Si

Summary

- **Control of the phonon propagation**
  Periodic → Aperiodic Si–SiGe multilayers
- **Realization of the electron crystal–phonon glass concept**
  Thermoelectric transport in oxide-embedded nanoparticles

- **Thermoelectric properties:**
  - $\sigma \approx 150$ S/cm, $S \approx 500$ µV/K, $\kappa_{\perp} < 5$ W/(Km)
    for highly doped Si–$Si_{1-x}Ge_x$ aperiodic multilayers
  - $\sigma \approx 100$ S/cm, $S > 600$ µV/K, $\kappa \approx 1$ W/(Km)
    for Si-based 0–3 composites

- **Figure of merit**
  $ZT > 1$ at 300 K for optimized Si-based thin films

Acknowledgments

- Martin Schade, Andreas Kipke, Frank Syrowatka, Frank Heyroth, Georg Schmidt (CMAT Halle)
- Matthias Stordeur (HTC Halle)

- BMBF WING project SiGe-TE 03X3541

GEFÖRDERT VOM
Bundesministerium für Bildung und Forschung

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Werkstoffinnovationen für Industrie und Gesellschaft

References

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