Thermal Conductivity and Interface Thermal Conductance of Phase Change Materials

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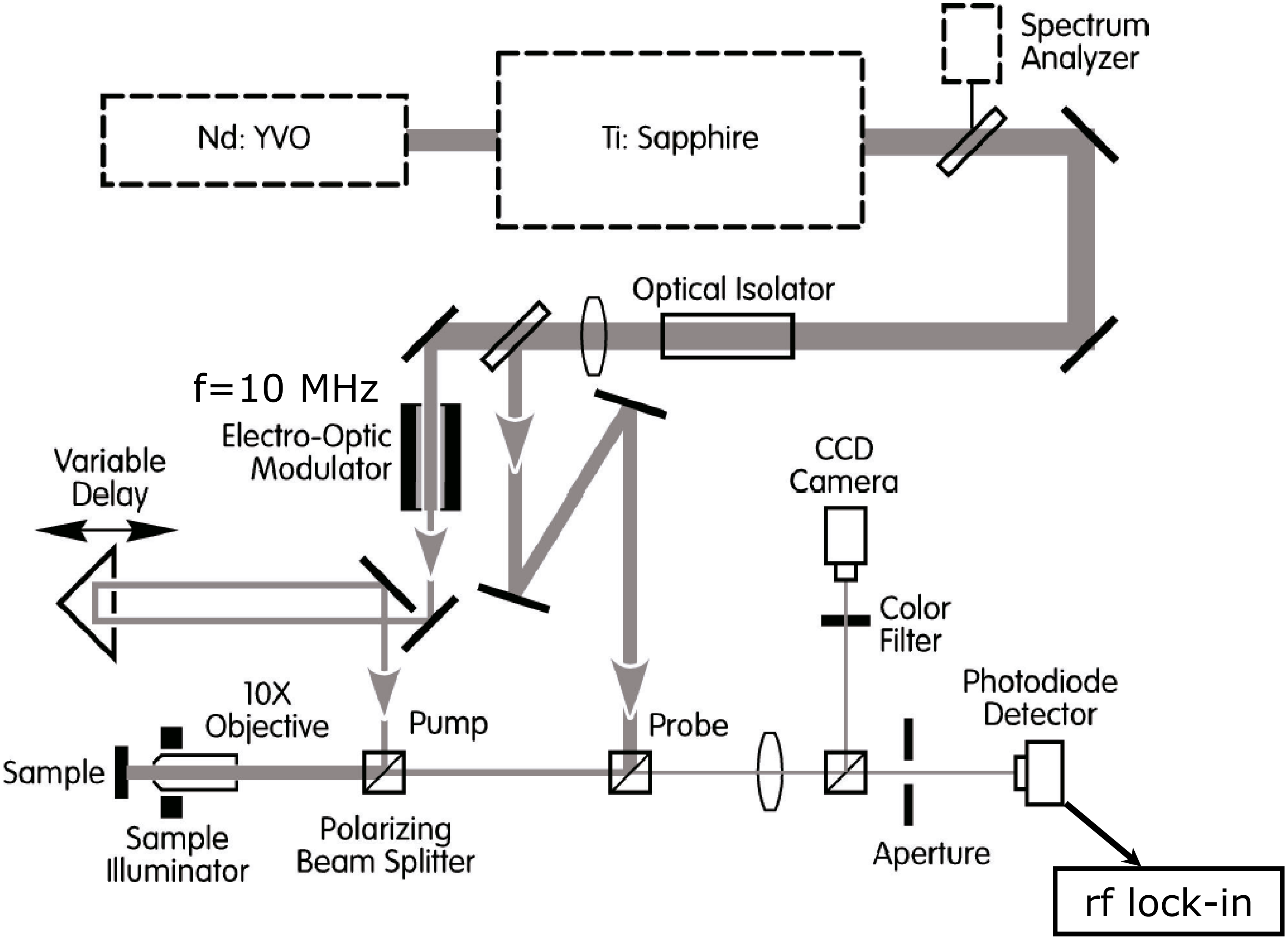
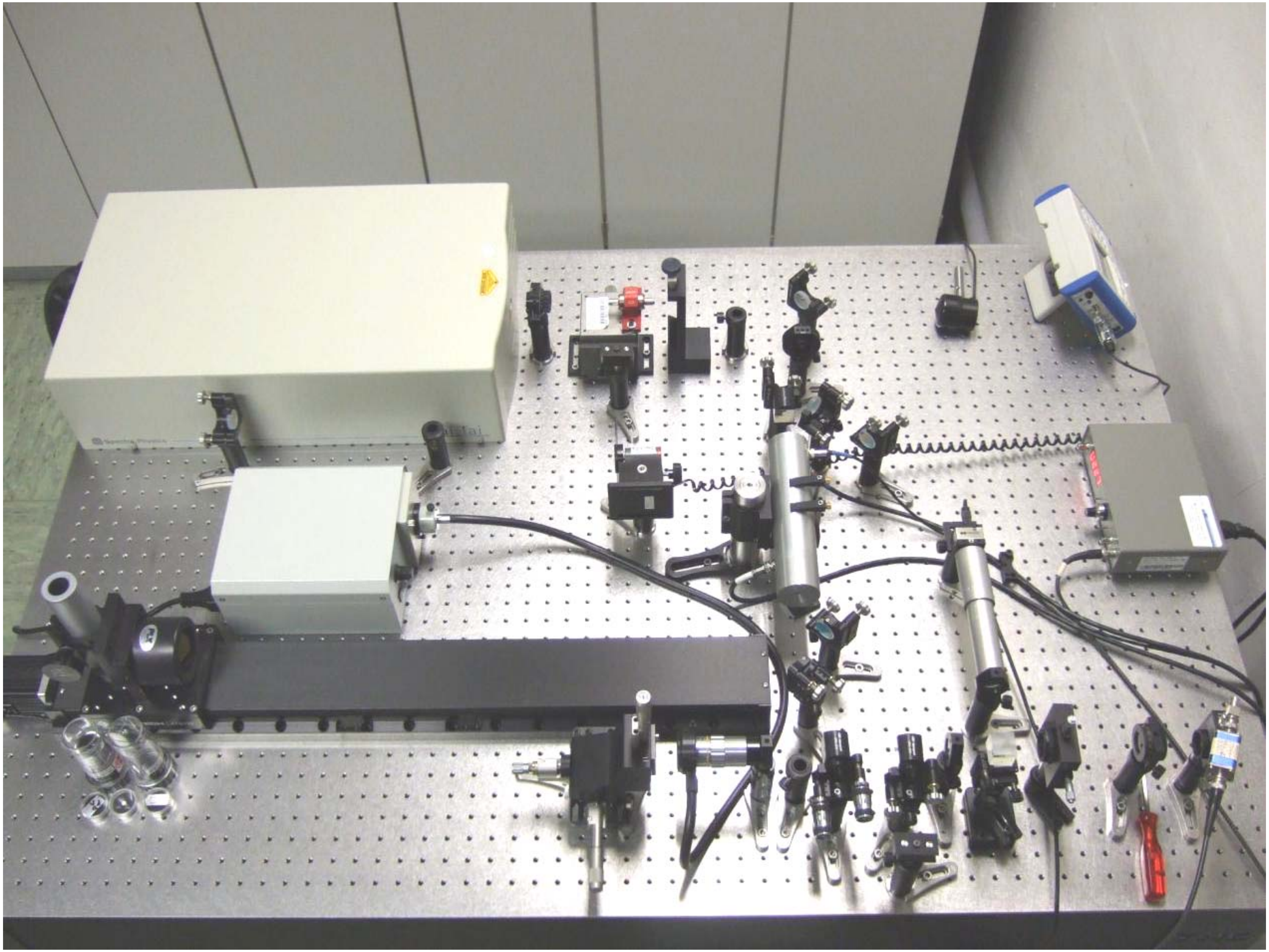
Outline

- Measurement: Modulated time-domain thermoreflectance (TDTR)
- Thermal conductivity: Phase change materials and the minimum thermal conductivity.
- Thermal conductance of interfaces with electrodes: Interfaces between highly dissimilar materials and anharmonic phonon transport.
- Controlling thermal conductance with thin interfacial layers: C_{60} films (demonstrated); disordered layered crystals WSe_2 (proposed).

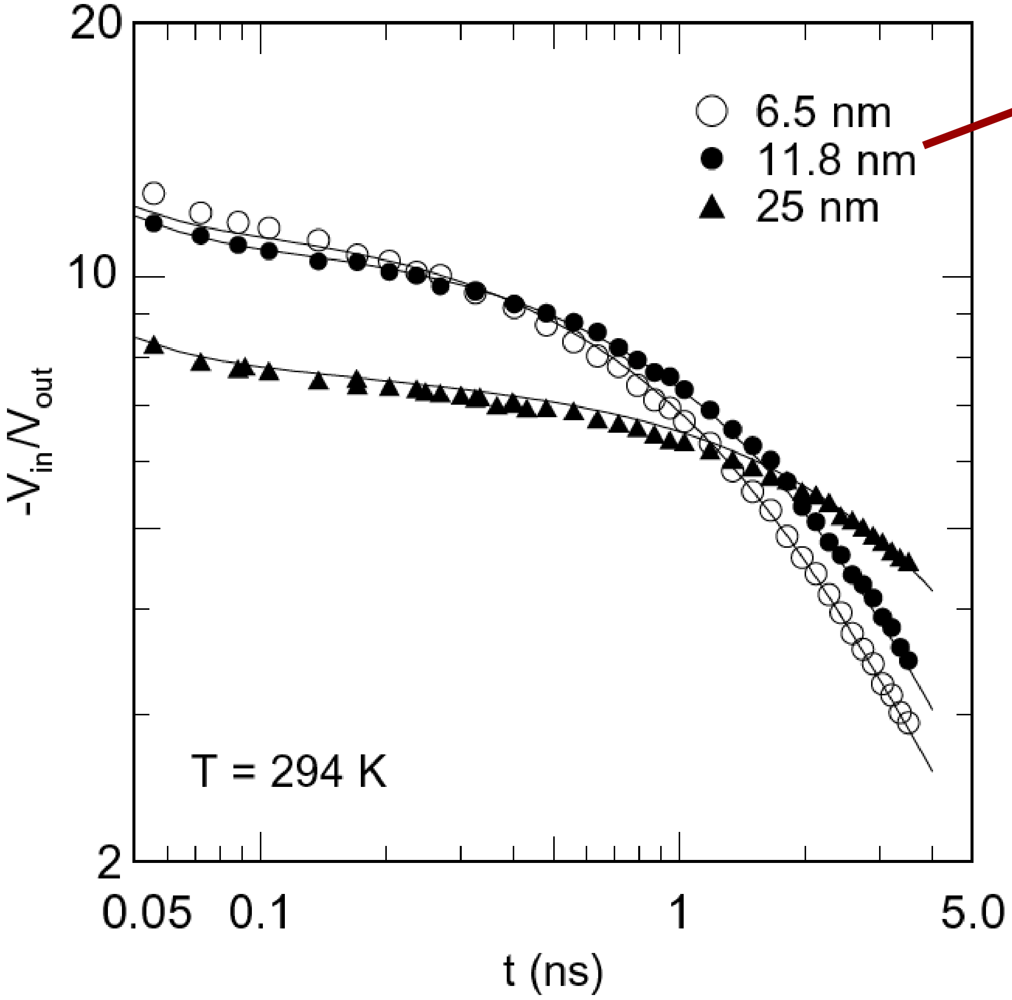
Modulated pump-probe apparatus

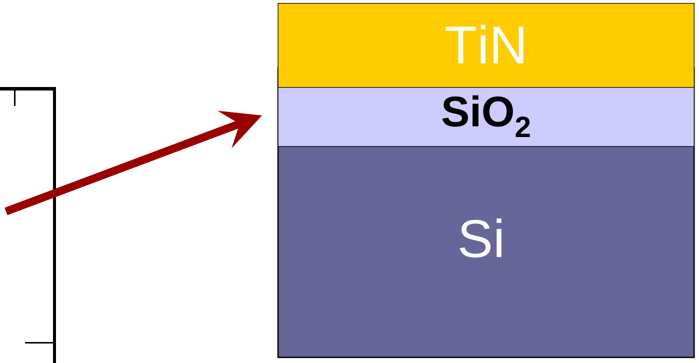


IPM system built January 7-8, 2008



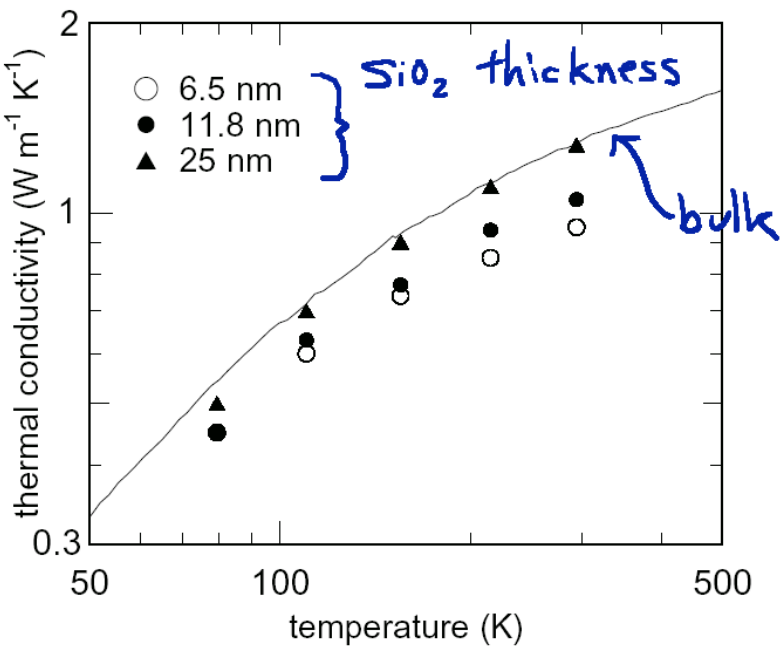
Time-domain Thermoreflectance (TDTR) data for TiN/SiO_2/Si

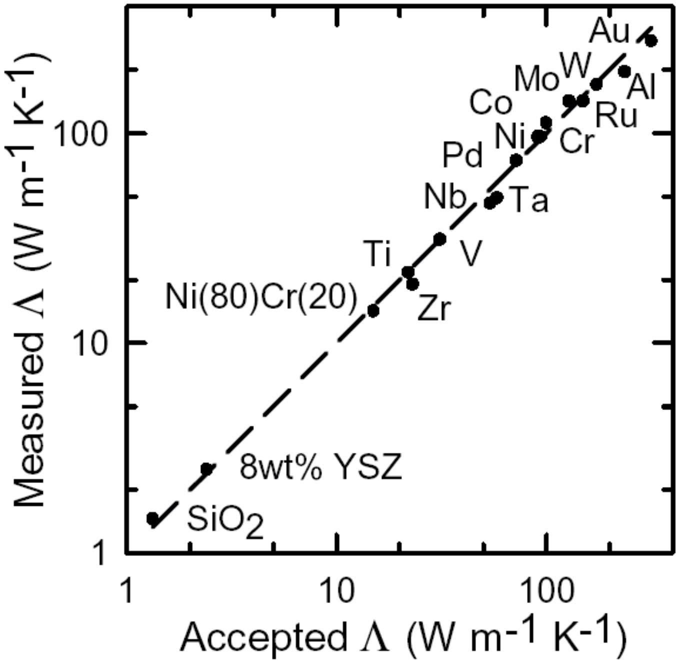




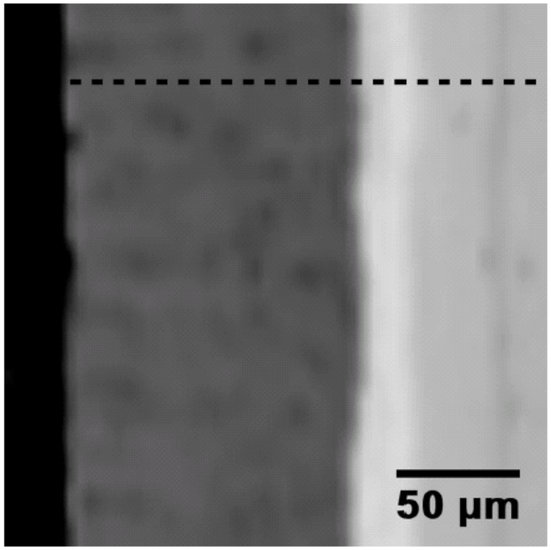
- reflectivity of a metal depends on temperature
- one free parameter: the "effective" thermal conductivity of the thermally grown SiO_2 layer (interfaces not modeled separately)

Flexible, convenient, and accurate technique...





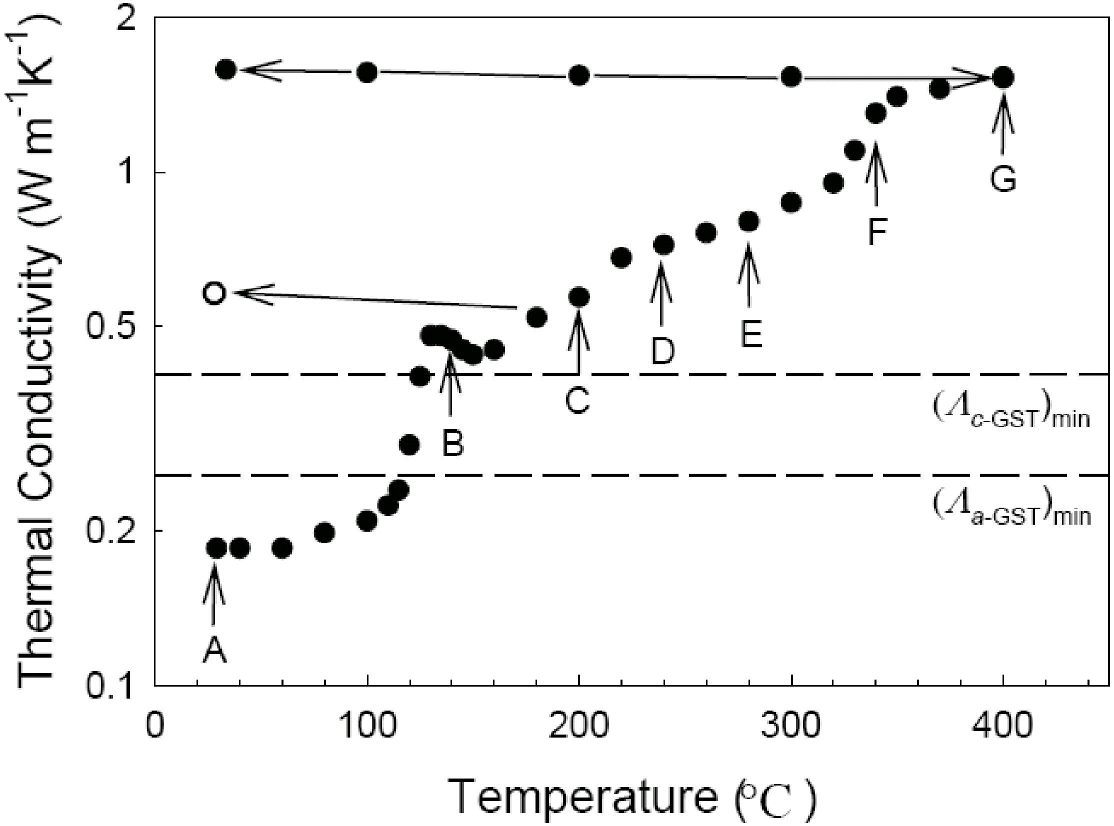
- ...with 3 micron resolution



thermal conductivity map of cross-section of thermal barrier coating, with J.-C. Zhao (GE)

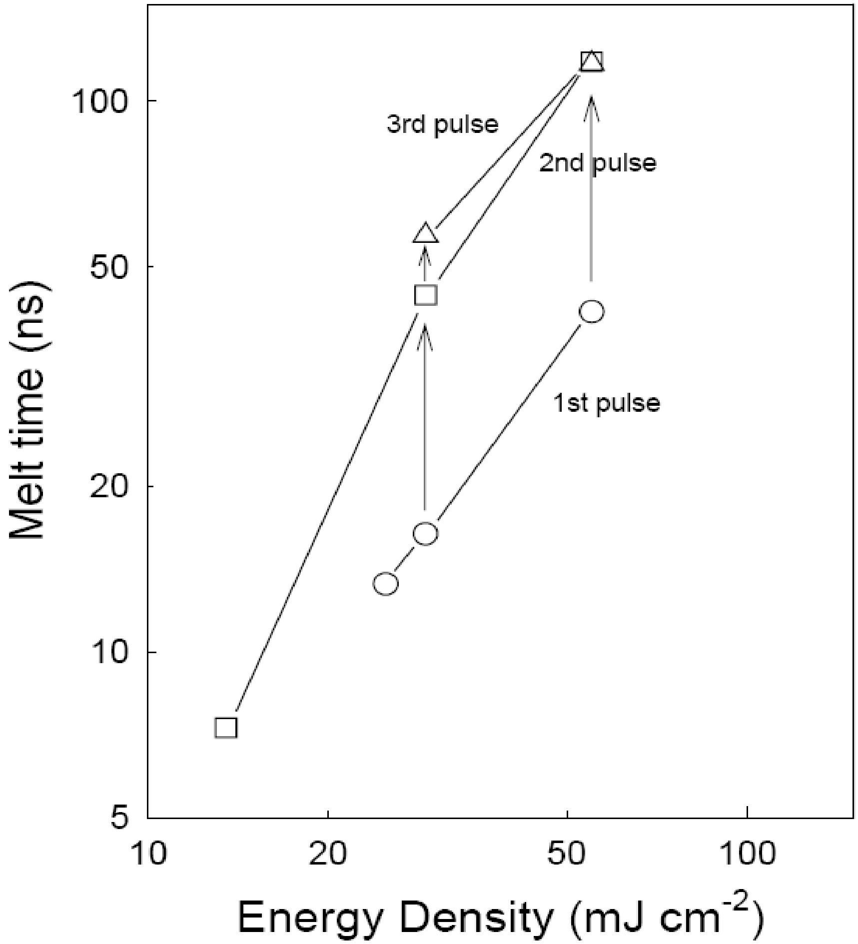
$Ge_2Sb_2Te_5$ during temperature ramp

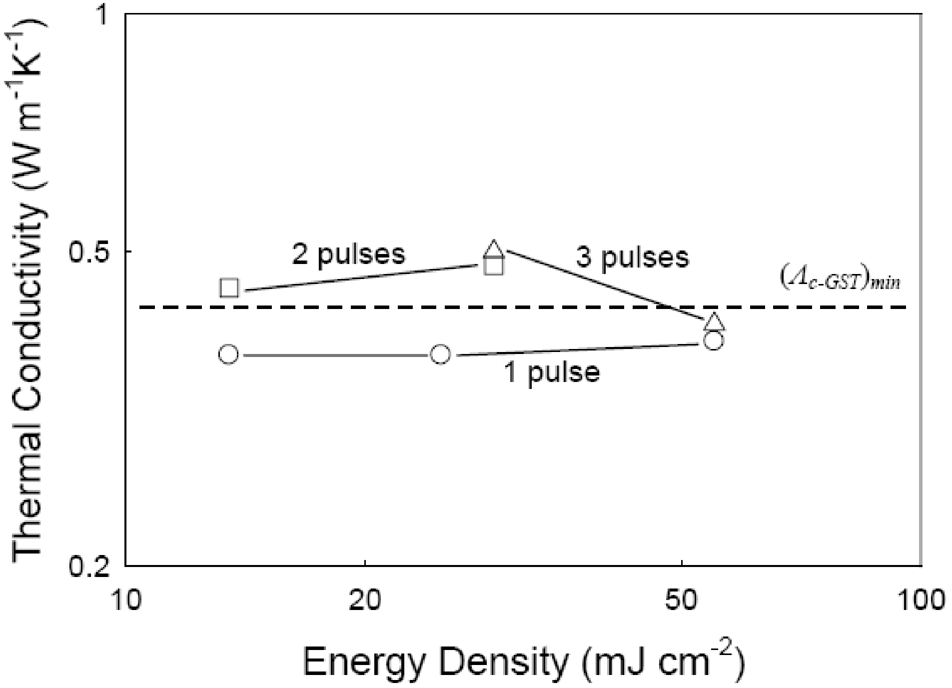
- Low conductivity in the cubic-phase (comparable to predicted Λ_{min}) increases modestly with annealing.



Cubic $Ge_2Sb_2Te_5$ formed by nsec laser pulse

- 523 nm, Q-switched doubled-YAG laser





Minimum thermal conductivity

- Both amorphous and "early" cubic phase have thermal conductivities comparable to the predicted minimum conductivity based on atomic density *n* and speeds of sound *v.*

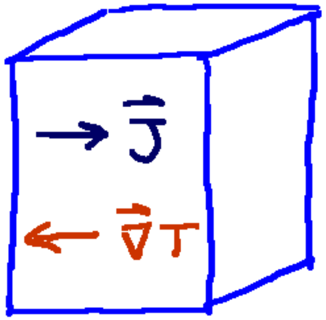
High T limit $$\Lambda_{\min} = \frac{1}{2}\left(\frac{\pi}{6}\right)^{1/3} k_B n^{2/3} (v_l + 2v_t)$$

- v_l measured directly by picosecond acoustics
- Assume $v_t = 0.6\ v_l$

Thermal conductivity and interface thermal conductance

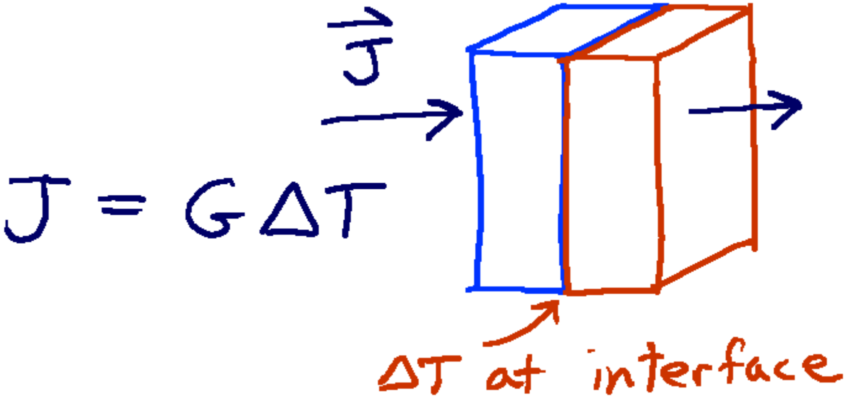
- Thermal conductivity Λ is a property of the continuum

$$\vec{J} = -\Lambda \vec{\nabla} T$$



$$\Lambda = \frac{1}{3 V k_B T^2} \int_0^\infty \langle \vec{j}(t) \cdot \vec{j}(0) \rangle \, dt$$

- Thermal conductance (per unit area) G is a property of an interface



$$J = G \Delta T$$

$$G = \frac{1}{A k_B T^2} \int_0^\infty \langle q(t) q(0) \rangle \, dt$$

Interface thermal conductance between GST and electrodes

- Difficult to measure because thermal conductivities are small and, for c-GST, depends on thickness; see Reifenberg et al. (2007) and Lee et al. (2000).

- And hard to predict because analytical models do not include anharmonicity or details of the interface structure and bonding.

- high temperature limit of the *radiation limit*

$$G = \frac{\pi}{3} \frac{k_b \nu_{\max}^3}{v_D^2}$$

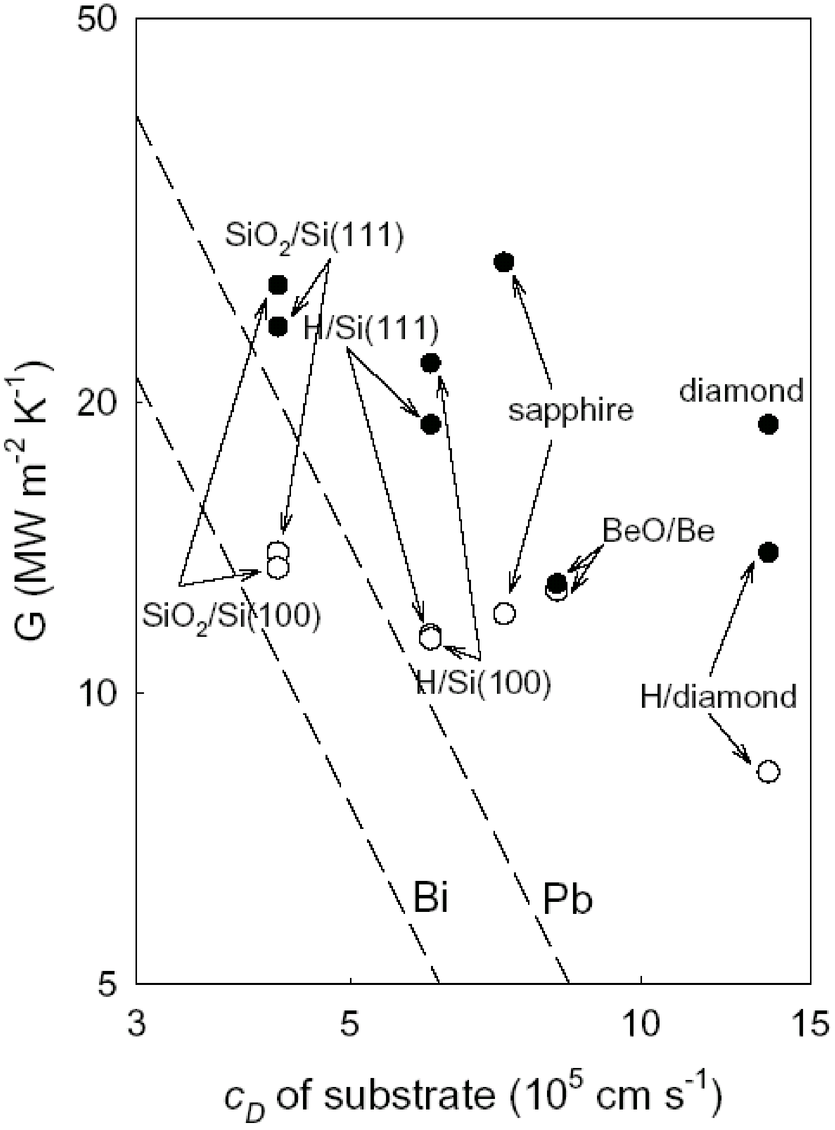
$\nu_{\max}$: vibrational cutoff frequency of material A

$(\nu_{\max} = 1.8$ THz for Bi, 2.23 THz for Pb)

$\mathrm{v_D}$: Debye velocity of material B

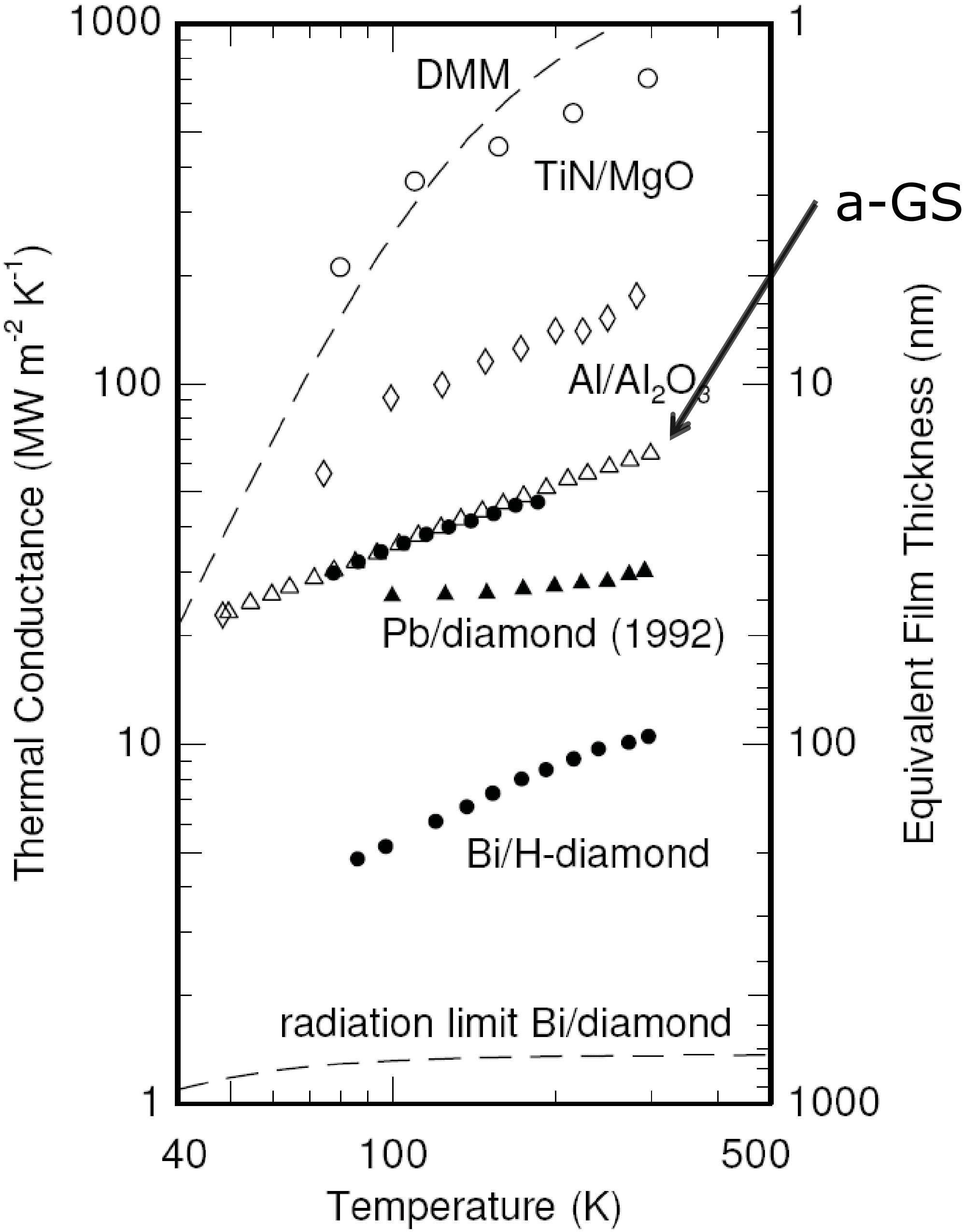
R. J. Stoner and H. J. Maris, *Phys.Rev.B* **48**, 22, 16373 (1993)

Room temperature thermal conductance



- Pb and Bi show similar behavior. Electron-phonon coupling is not an important channel.
- Weak dependence on Debye velocity of the substrate.
- For Pb and Bi, conductance always larger than predicted by a purely elastic process.

Interface thermal conductance: Factor of 60 range at room temperature



a-GST/ZnS:SiO$_2$ Lee et al. (2000)

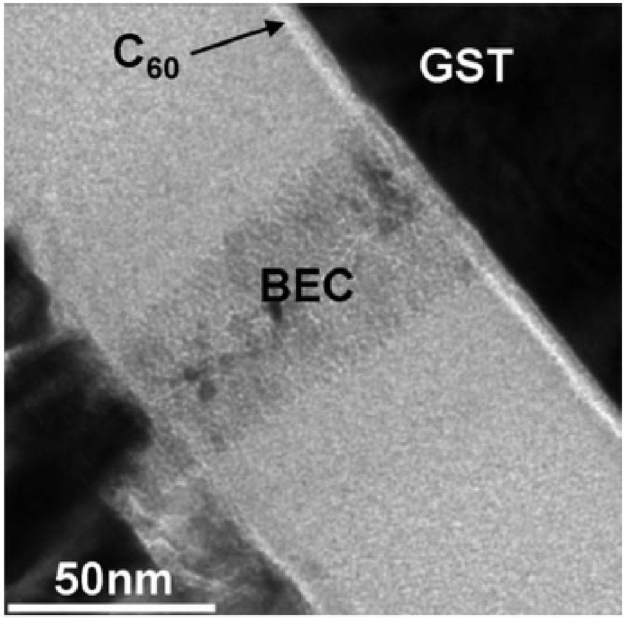
$L = \Lambda/G$

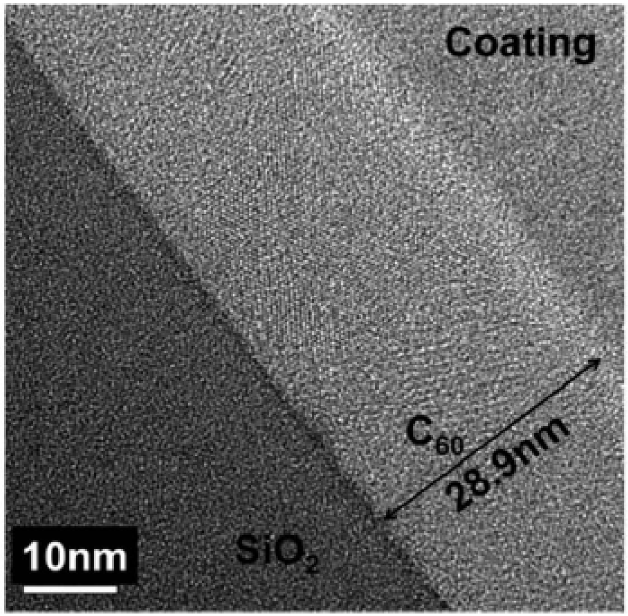
$\Lambda = 1$ W m^{-1} K^{-1}

Bottom line…

- Thermal conductance of $Ge_2Sb_2Te_5$ /nitride interfaces is not known precisely. Limited data and analogy to Pb interfaces suggests $G \approx 25$ MW m^{-2} K^{-1} at room temperature.
- Kapitza length $L = \Lambda/G \approx 10$ nm for a-$Ge_2Sb_2Te_5$
- Not yet measured but G will probably increase significantly with temperature.
- For liquid (metallic) $Ge_2Sb_2Te_5$, conductance will become large because of electronic thermal transport.

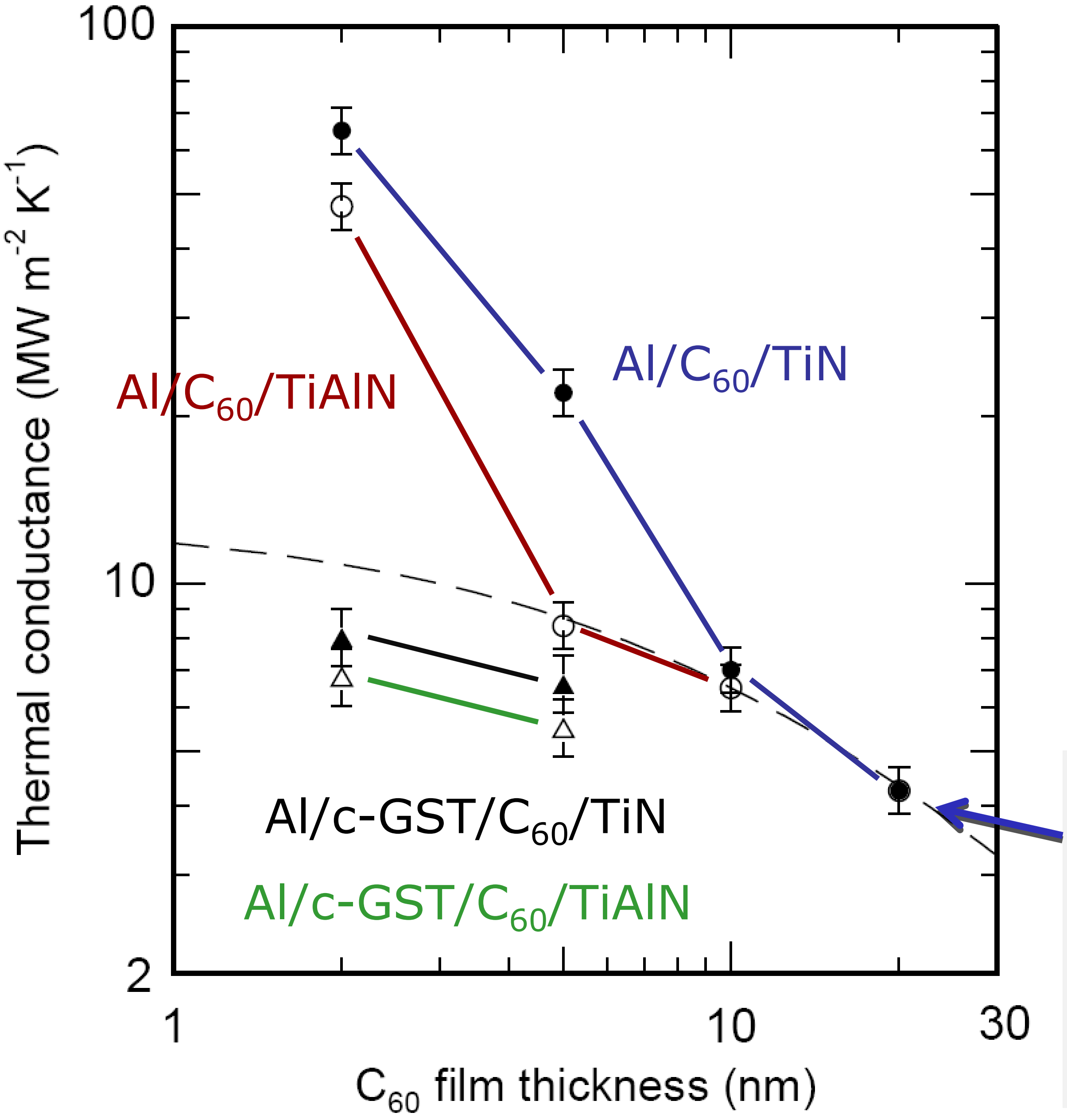
C_{60} fullerene as thermal insulation

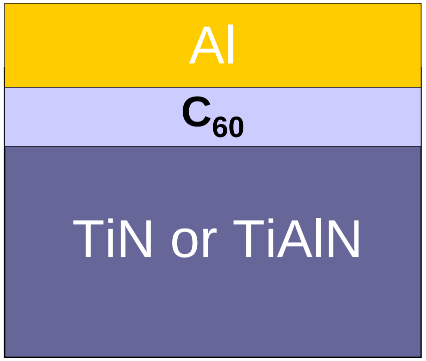


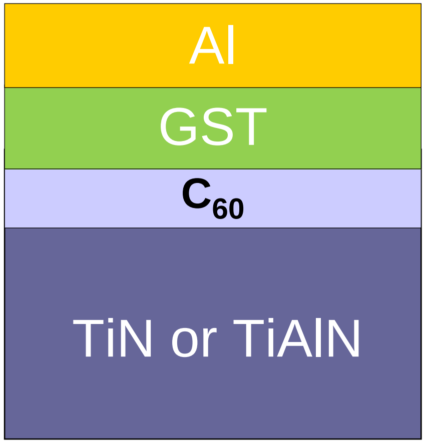


- Evaporate C_{60} on TiN or TiAlN back-electrode contacts
- Add $Ge_2Sb_2Te_5$ layer (or not)
- Coat with Al for thermal transport measurements by time-domain thermoreflectance

C_{60} fullerene as thermal insulation







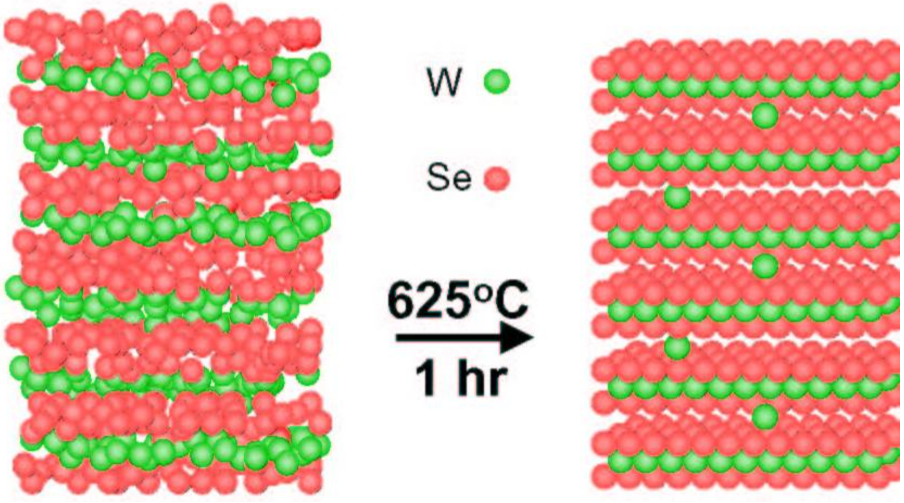
Fit gives interface conductance and conductivity of C_{60}

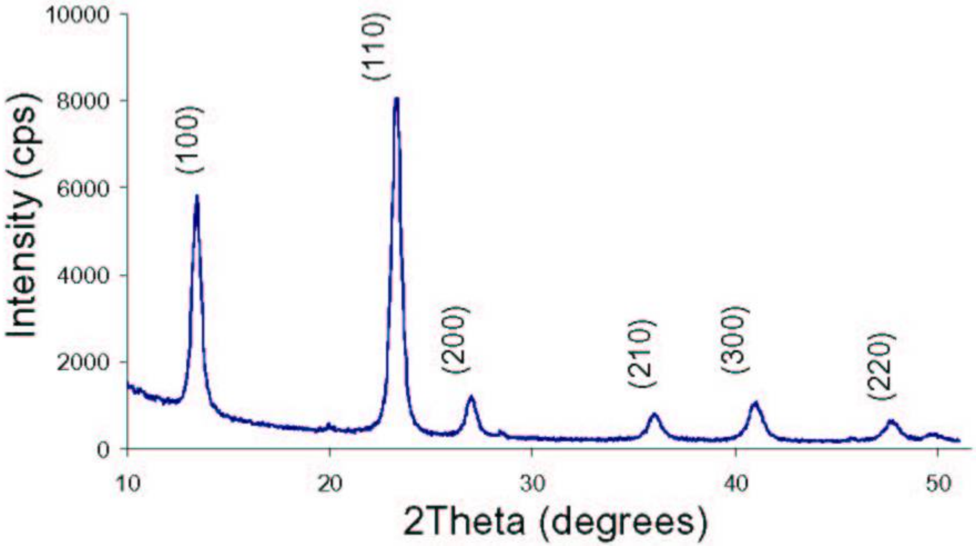
$G=13$ MW m^{-2} K^{-1}

$\Lambda = 0.13$ W m^{-1} K^{-1}

Layered disordered crystals: WSe$_2$ by “modulated elemental reactants”

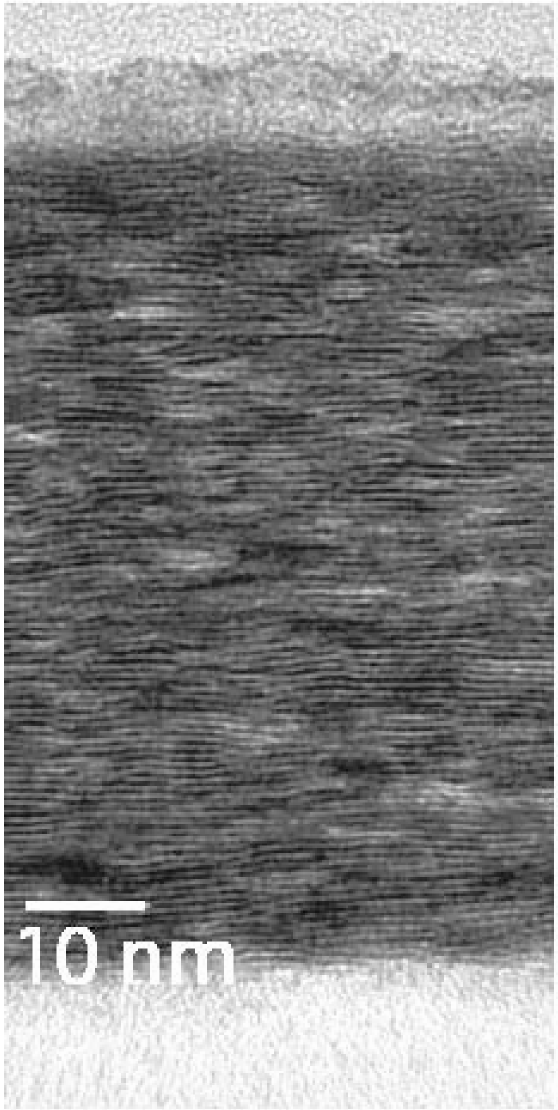
- Deposit W and Se layers at room temperature on Si substrates
- Anneal to remove excess Se and improve crystallinity
- Characterize by RBS, x-ray diffraction (lab sources and Advanced Photon Source) and TEM

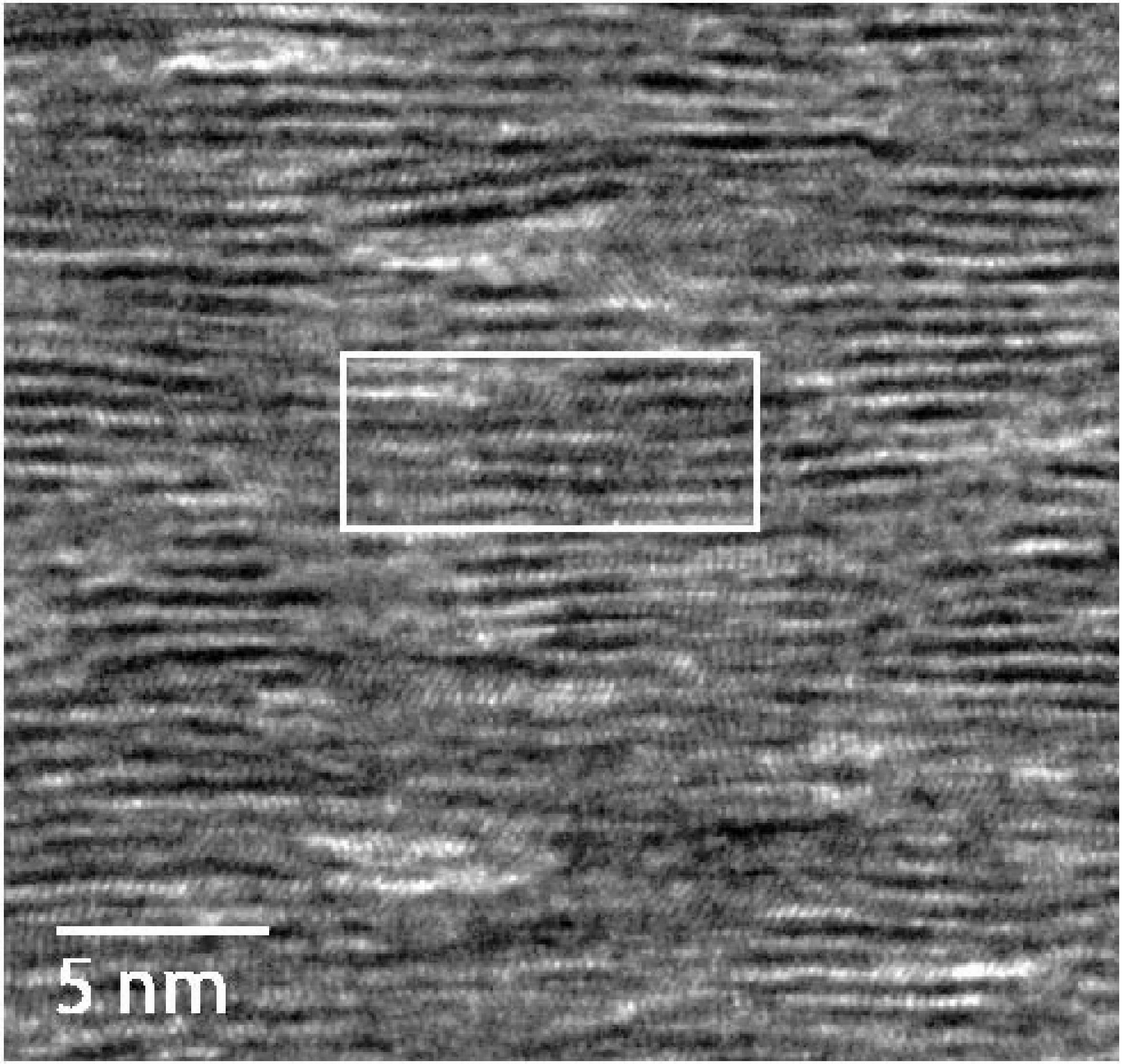




David Johnson group, U. Oregon

Cross-sectional TEM of 60 nm thick WSe_2



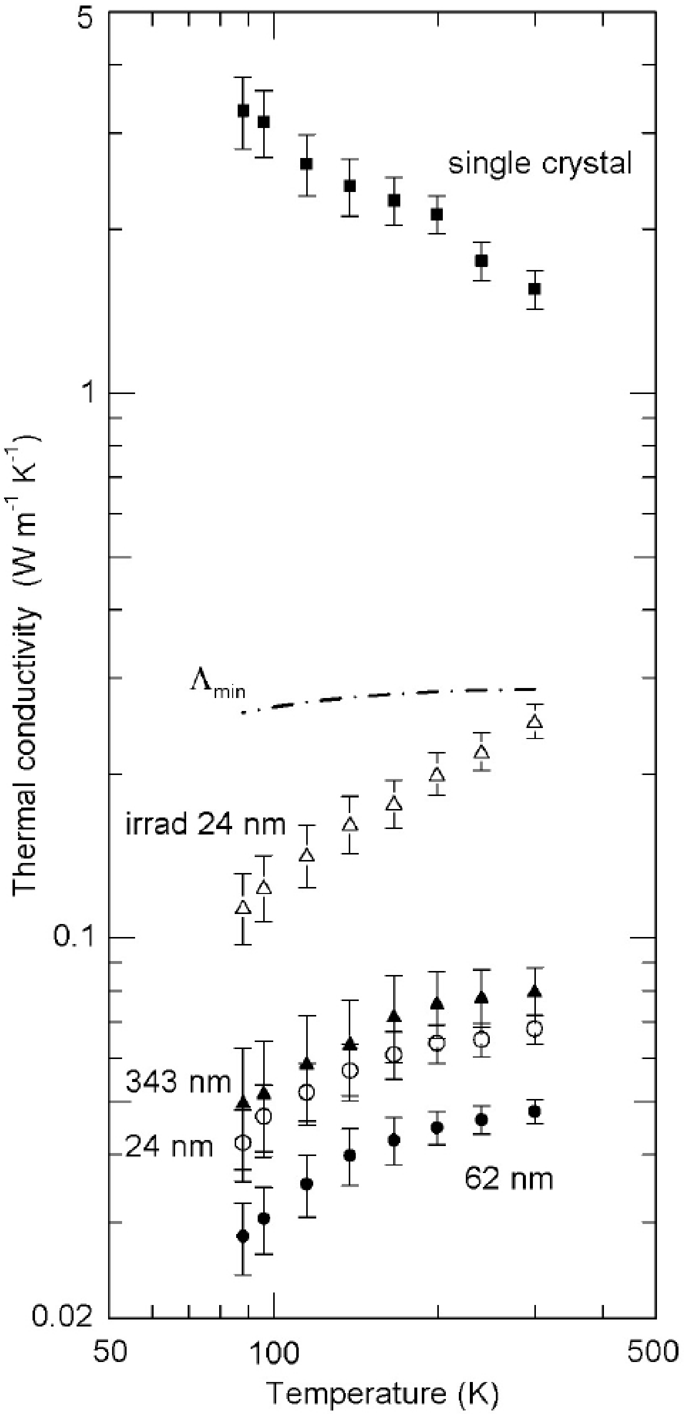


Seongwon Kim and Jian Min Zuo

Thermal conductivity of WSe_2

- 60 nm film has the lowest thermal conductivity ever observed in a fully dense solid. Only twice the thermal conductivity of air.
- A factor of 6 less than the calculated amorphous limit for this material.

Chiritescu et al. Science (2006)



Conclusions

- Thermal conductivity of amorphous and “early” cubic phase and laser crystallized cubic phase are all comparable to the predicted minimum thermal conductivity ➔ strong disorder in the crystal
- Thermal conductance of interfaces with nitride electrodes is equivalent to ≈10 nm thick layer of amorphous GST, decreases with thickness.
- C_{60} layer provides thermal resistance equivalent to ≈20 nm thick layer of amorphous GST
- Could, in principle produce the same thermal resistance with a 5 nm thick layer of a *disordered layered crystal* such as WSe_2.