Lattice Vibrations in Amorphous Thin Films: A Path to Nanocomposite Based Energy Conversion?

Bruce White

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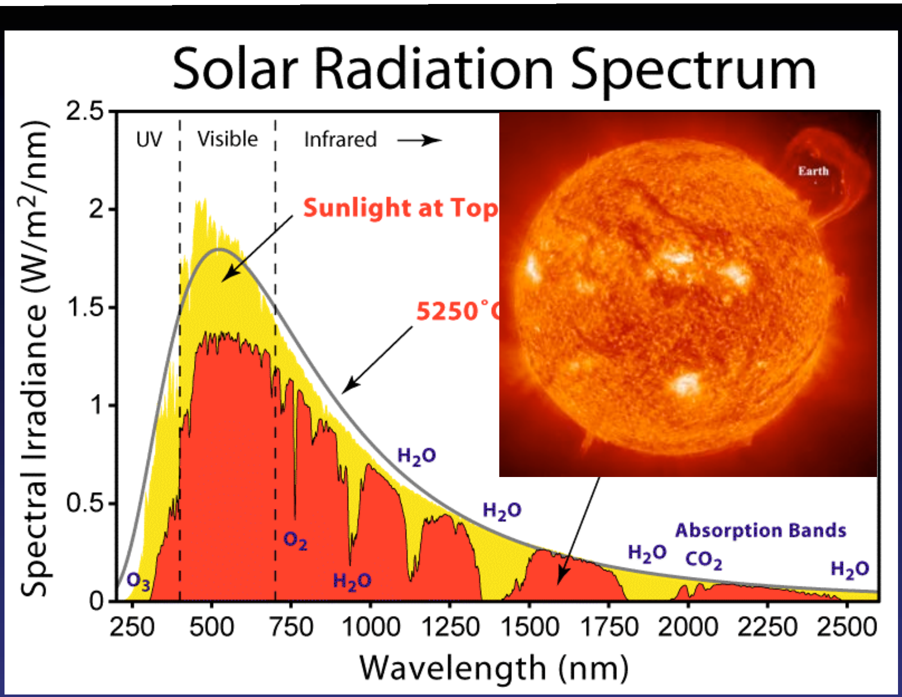

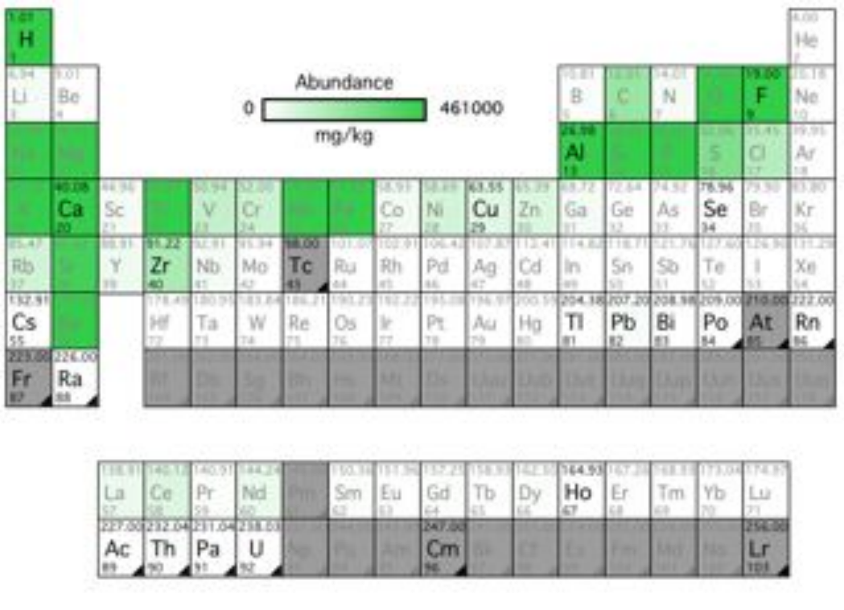

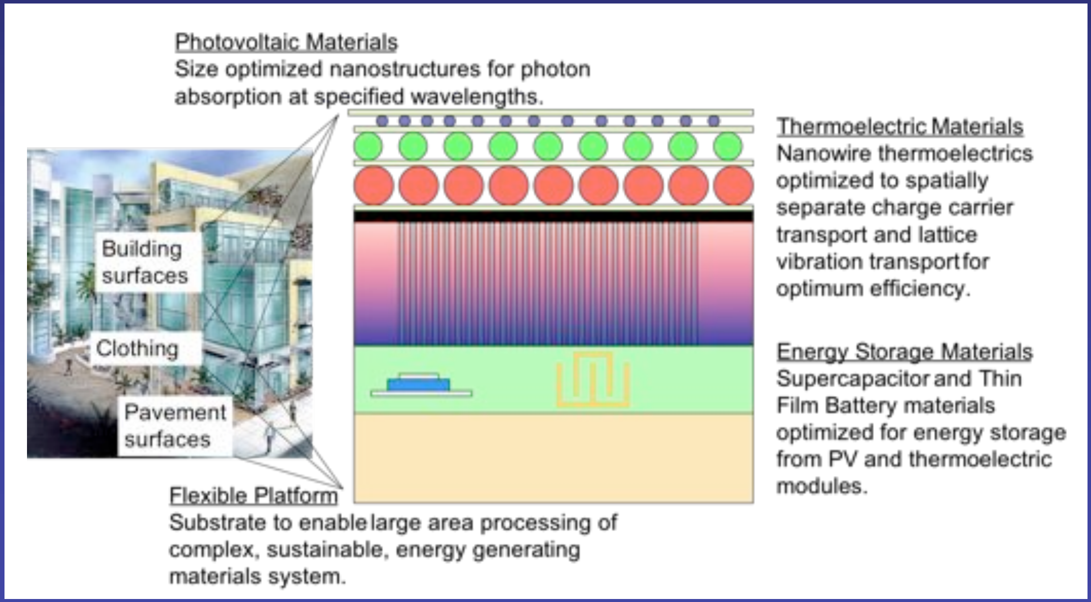

Photovoltaic Materials
Size optimized nanostructures for photon absorption at specified wavelengths.

Thermoelectric Materials
Nanowire thermoelectrics optimized to spatially separate charge carrier transport and lattice vibration transport for optimum efficiency.

Energy Storage Materials
Supercapacitor and Thin Film Battery materials optimized for energy storage from PV and thermoelectric modules.

Flexible Platform
Substrate to enable large area processing of complex, sustainable, energy generating materials system.

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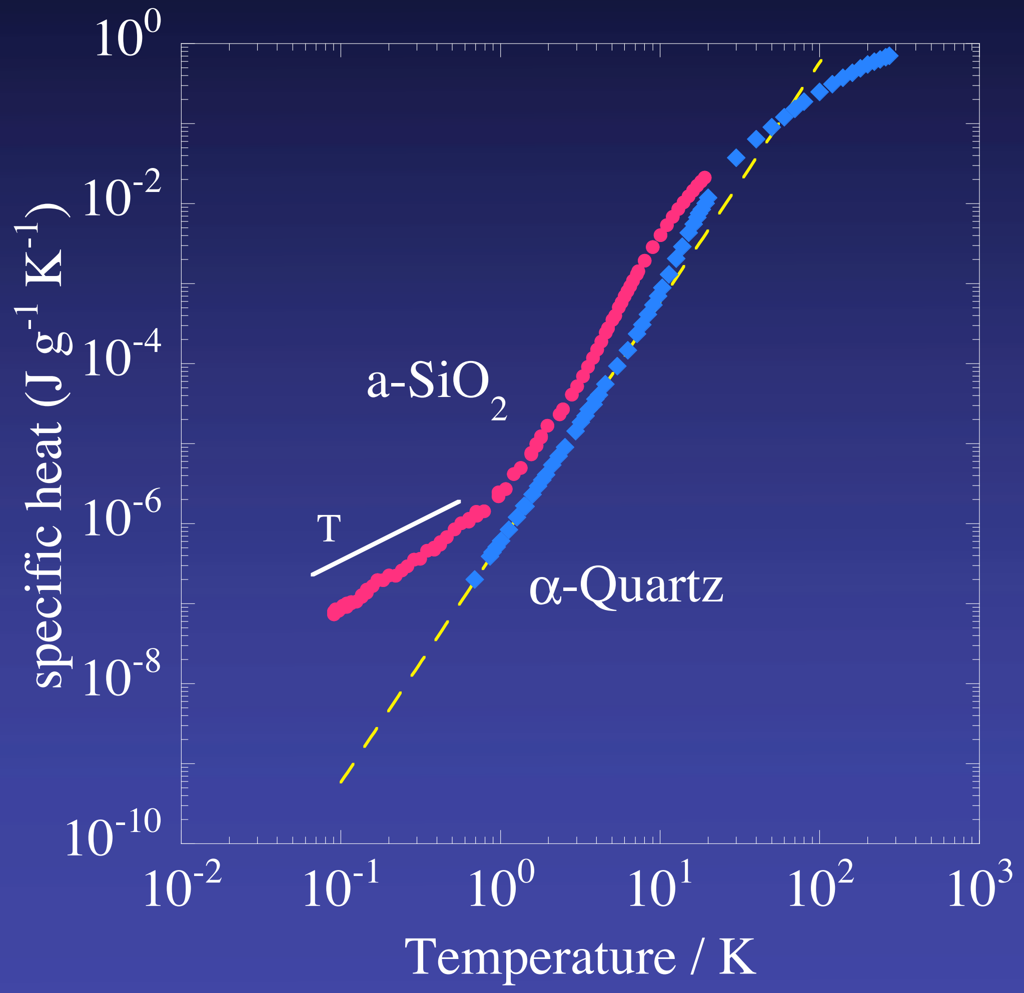
Anomalous Heat Capacity of Amorphous Solids



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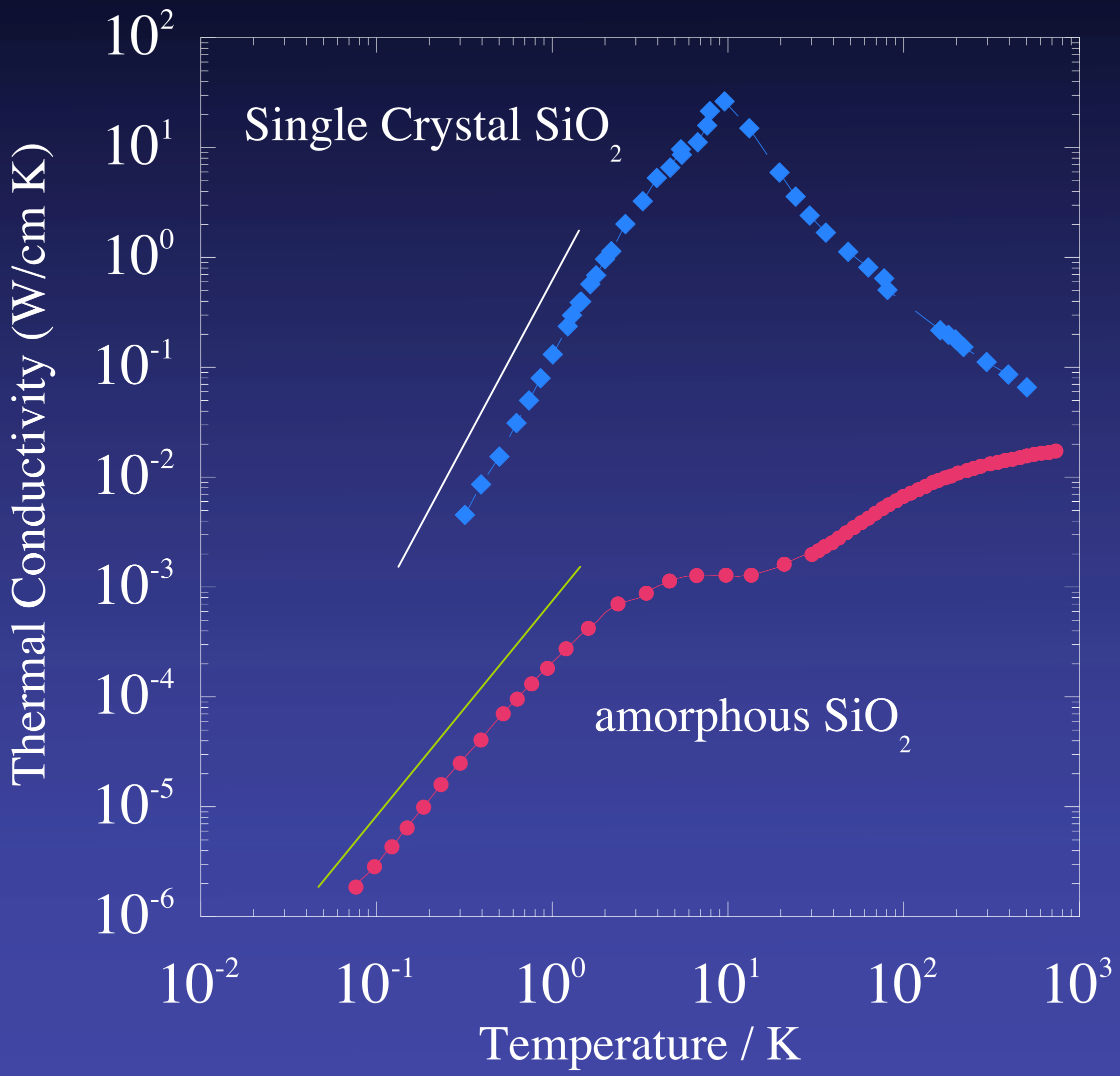
Anomalous Low T Thermal Conductivity



$$\kappa_{phonons} = \frac{1}{3} C_v v l$$

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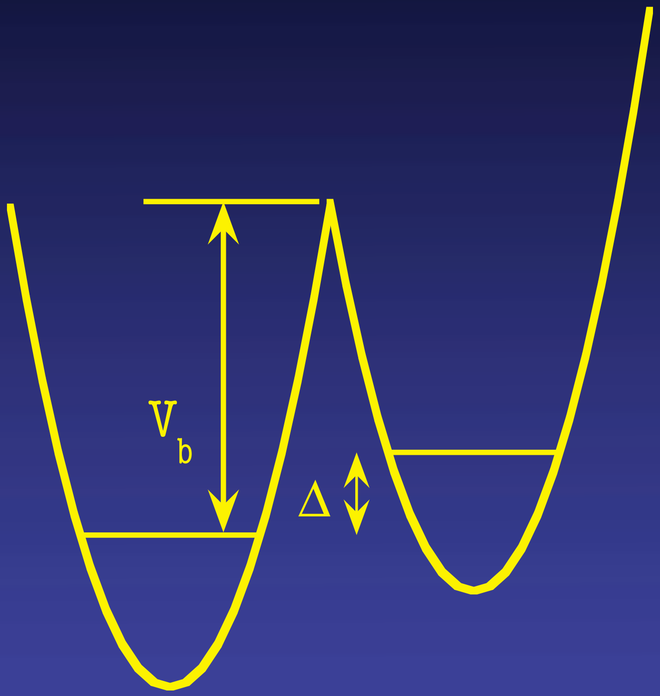
Tunneling Model



$$\Delta_o = \hbar \omega_o e^{-\lambda}$$

$$P(\lambda, \Delta) = \bar{P}$$

$$C_p = \frac{\pi^2 k_B^2}{12\rho} \bar{P} T \ln\left(\frac{4t}{\tau_{\mathrm{min}}}\right)$$

$$\Lambda = \frac{k_B^3}{6\pi\hbar^2} \frac{\rho \mathrm{v}_\mathrm{t}^2}{\bar{P}\gamma_t^2} \frac{1}{\mathrm{v}_t} \left(2 + \frac{\gamma_t^2 \mathrm{v}_\mathrm{l}}{\gamma_\mathrm{t}^2 \mathrm{v}_\mathrm{t}}\right) T^2$$

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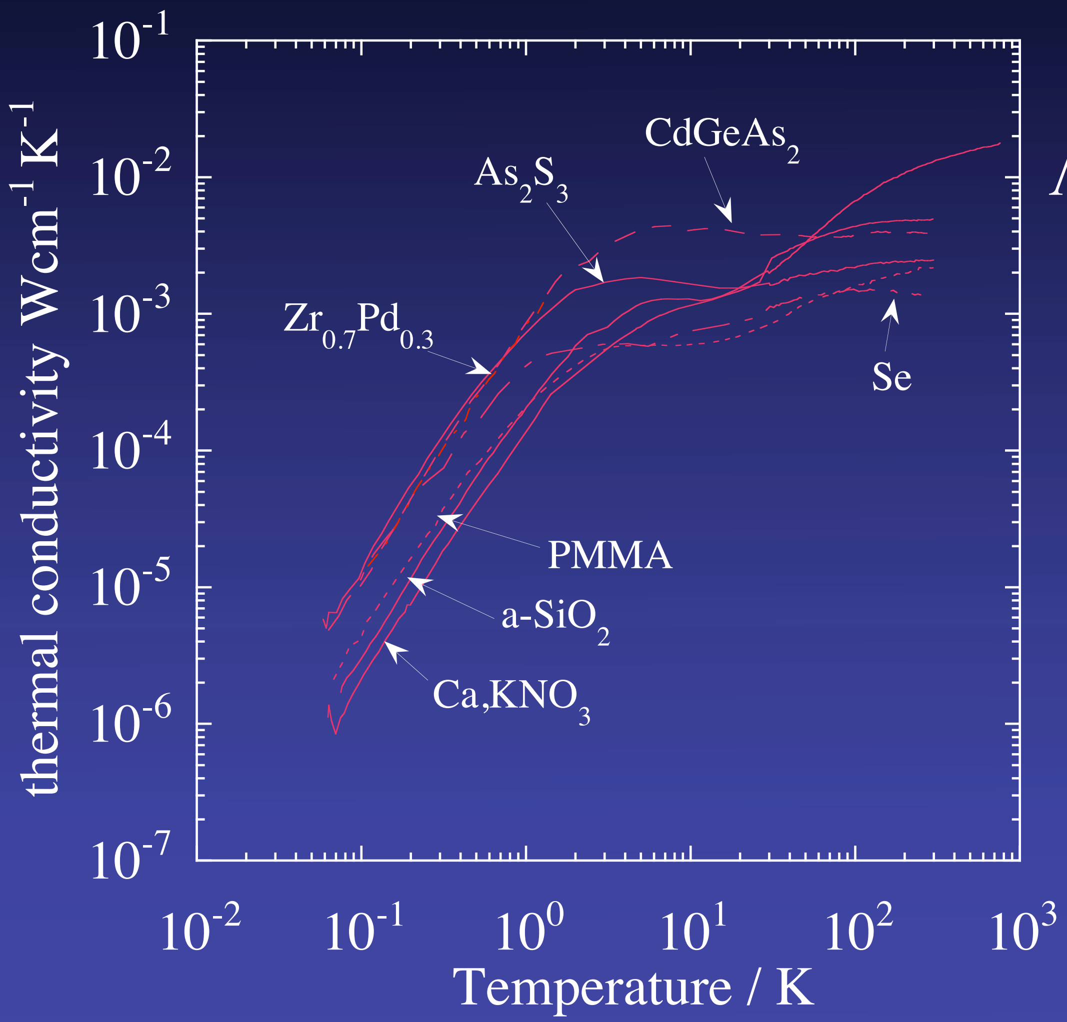
Universality in Glassy Lattice Vibrations

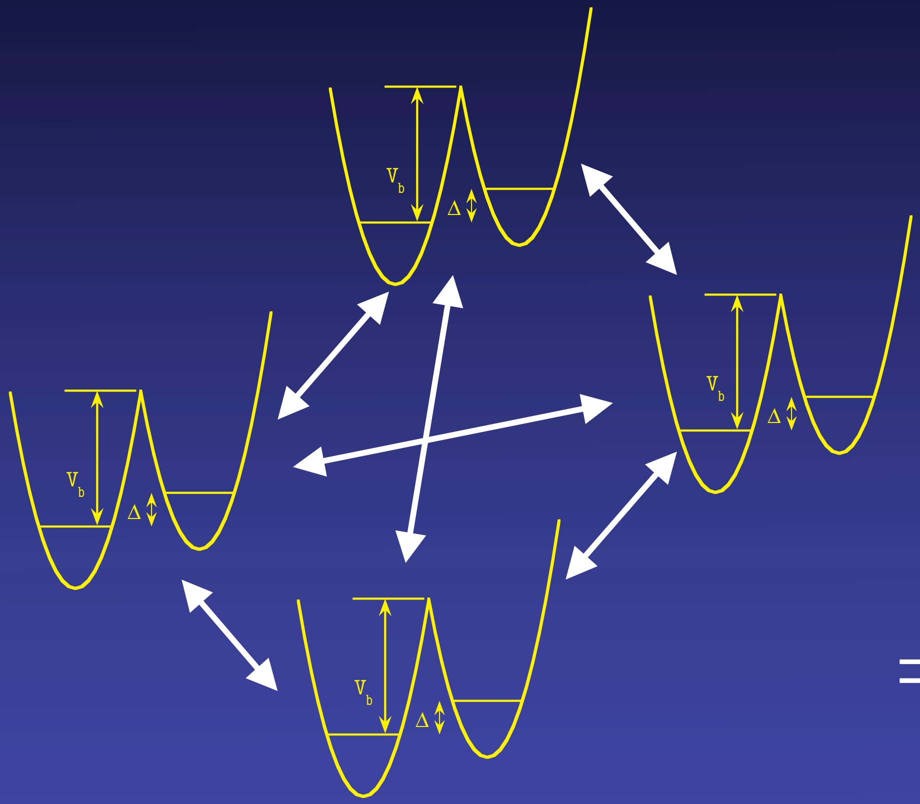


$$\Lambda = \frac{k_B^3}{6\pi\hbar^2}\frac{\rho v_t^2}{\bar{P}\gamma_t^2}\frac{1}{v_t}\left(2+\frac{\gamma_t^2 v_l}{\gamma_t^2 v_t}\right)T^2$$

$\frac{\bar{P}\gamma_t^2}{\rho v_t^2}$ constant within a factor 3 for all amorphous solids!

Why???

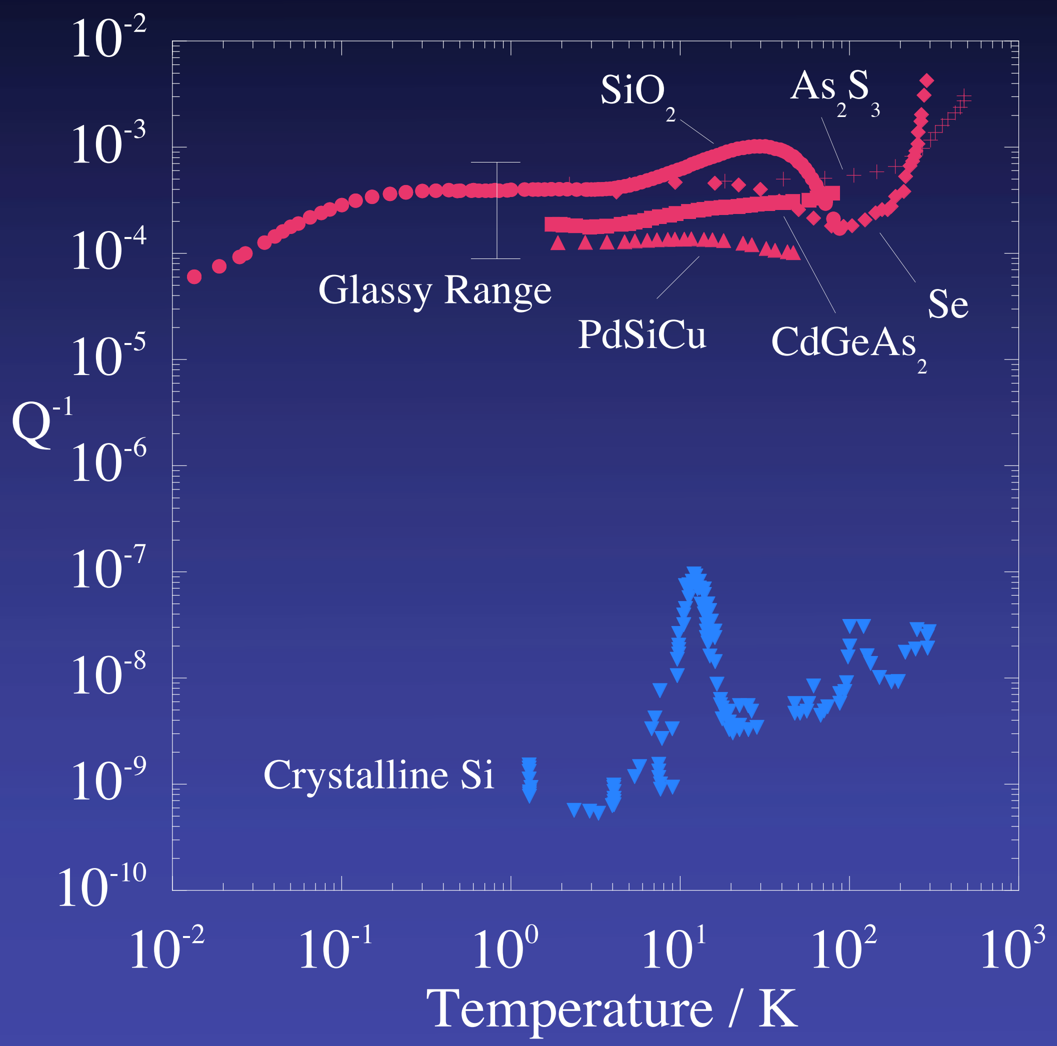
Source of Universality: Interactions between tunneling states?



$$\overline{P} \propto \frac{c_{44}}{\gamma_t^2}$$

$$\Rightarrow \frac{\overline{P}\gamma_t^2}{c_{44}} = const. = \frac{\overline{P}\gamma_t^2}{\rho v_t^2}$$

Universality in Elastic Properties



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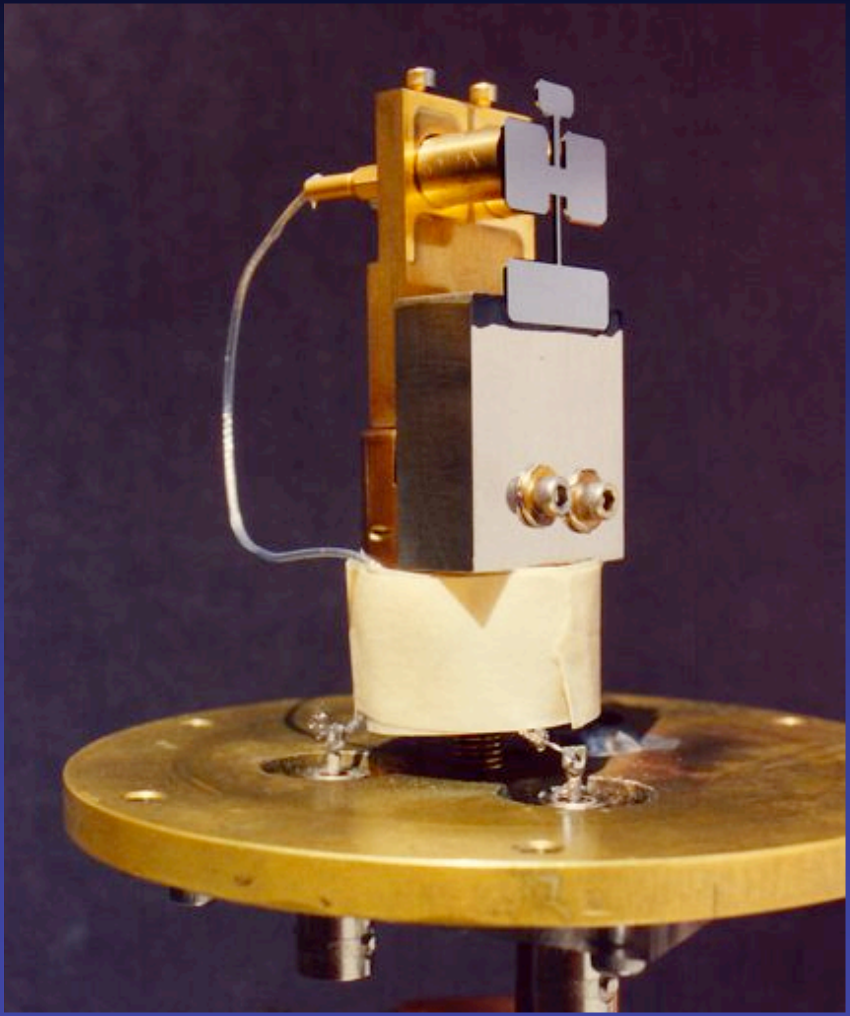
Elastic Properties of Thin Films



$$Q^{-1} = \frac{1}{2\pi}\left(\frac{\Delta E_{sub}}{E_{sub}} + \frac{E_{film}}{E_{sub}}\frac{\Delta E_{film}}{E_{film}}\right) = Q^{-1}_{sub} + \frac{E_{film}}{E_{sub}}Q^{-1}_{film}$$

$$Q^{-1} = Q^{-1}_{sub} + \frac{3t_{film}G_{film}}{t_{sub}G_{sub}}Q^{-1}_{film}$$

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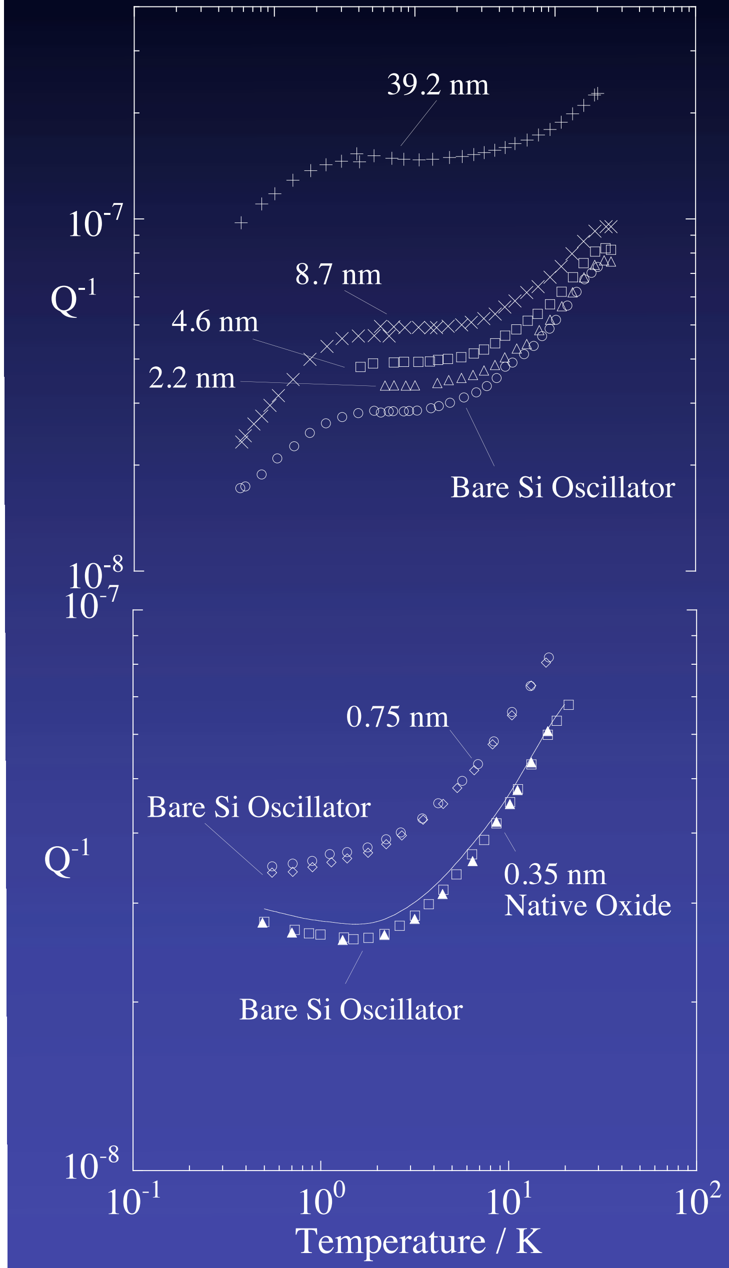
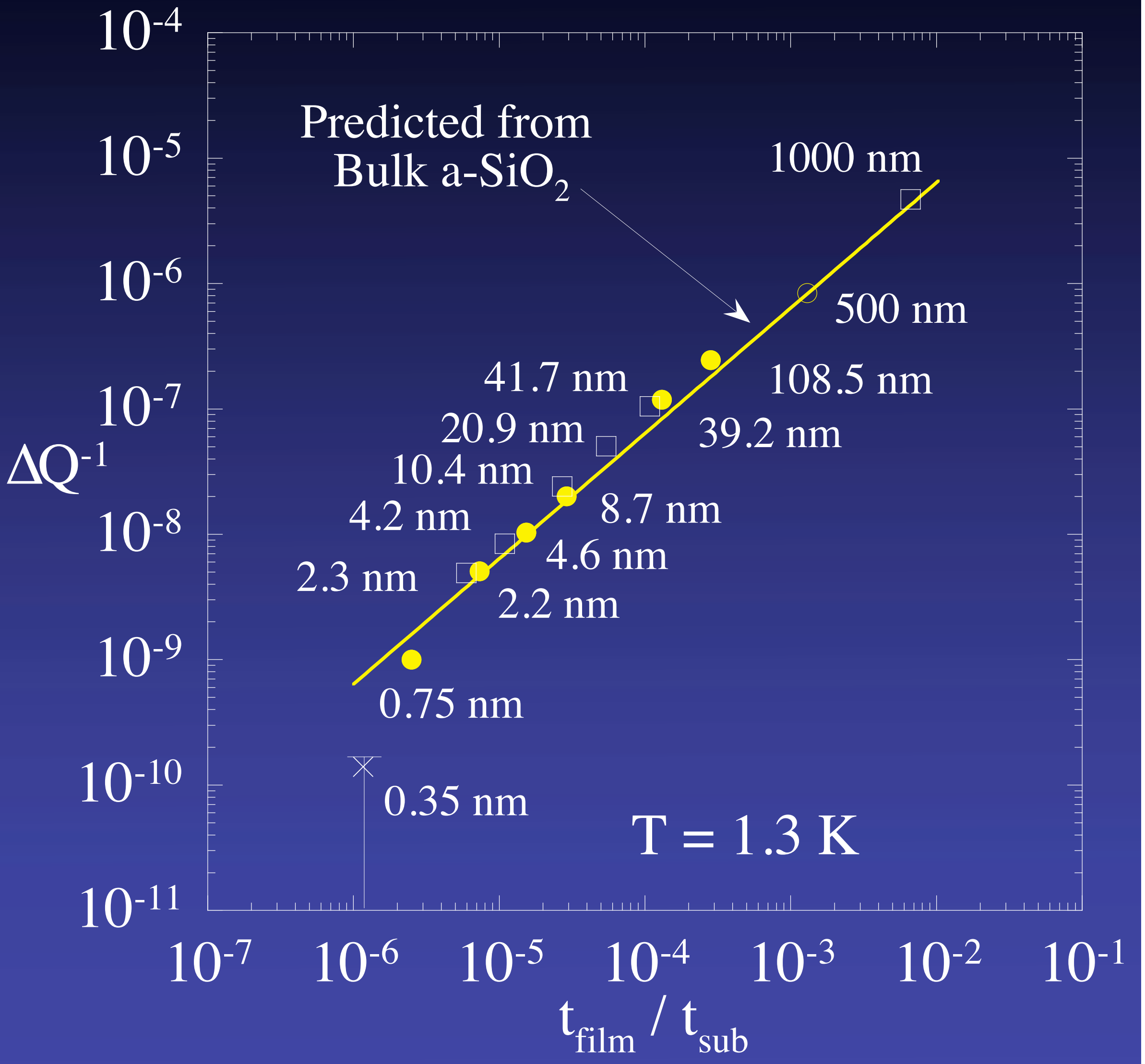
Ultra Thin SiO_2 Internal Friction





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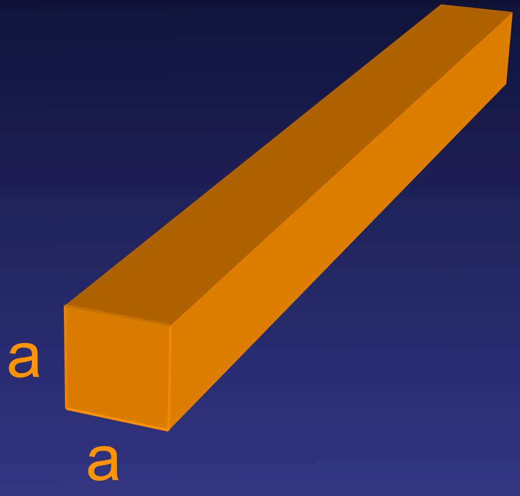
Thermoelectric Nanostructures

$$ZT = \frac{S^2 \sigma T}{\kappa_{ph} + \kappa_{el}}$$

$$\varepsilon\left(k_x, k_y\right) = \frac{\hbar^2 k_x^2}{2m_x} + \frac{\hbar^2 \pi^2}{2m_y a^2} + \frac{\hbar^2 \pi^2}{2m_z a^2}$$

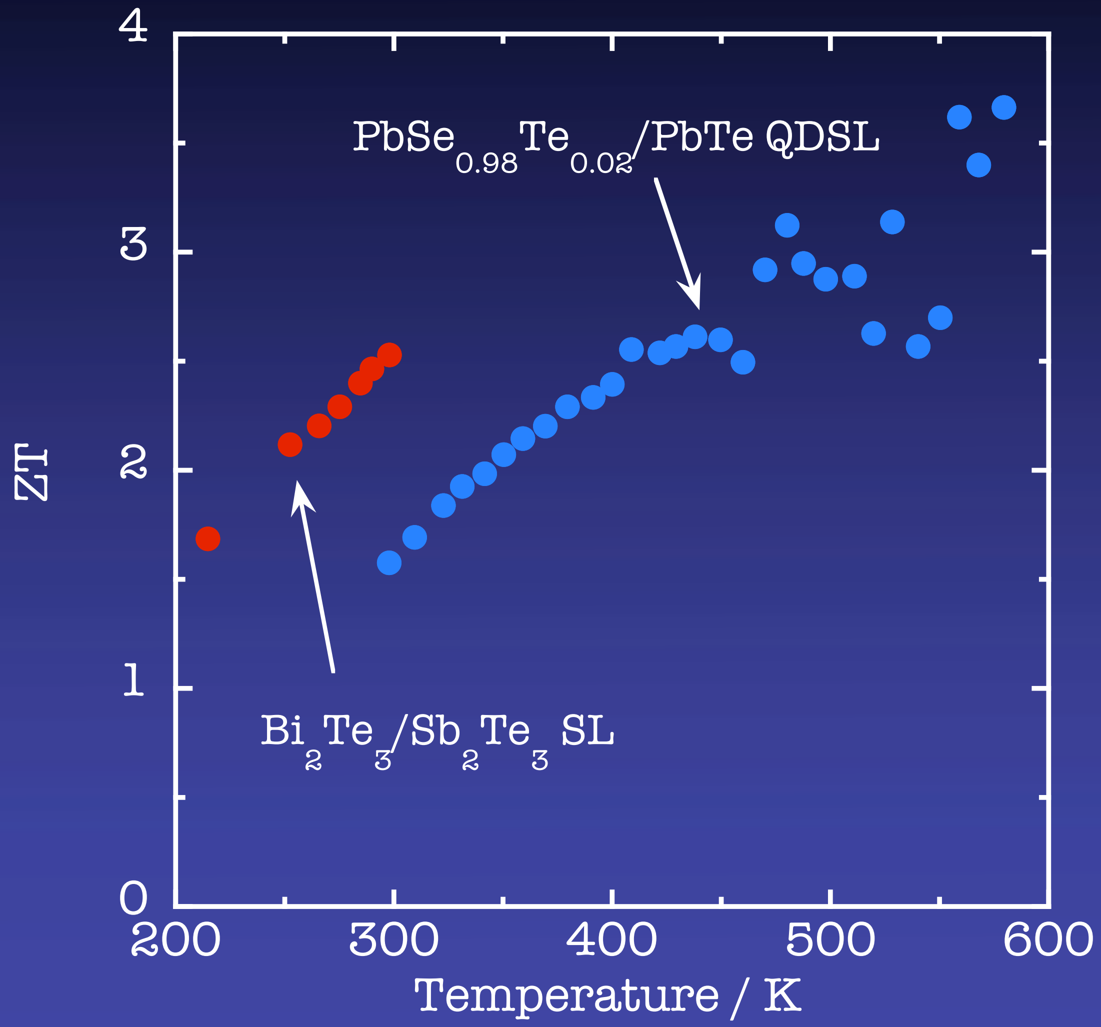
$$ZT = \frac{\frac{1}{2}\left(\frac{3F_{1/2}}{F_{-1/2}} - \eta\right)^2 F_{-1/2}}{\frac{1}{B} + \frac{5}{2}F_{3/2} - \frac{9F_{1/2}^2}{2F_{-1/2}}}$$

$$B = \frac{2}{\pi a^2}\left(\frac{2k_B T}{\hbar^2}\right)^{1/2} \frac{k_B^2 T m_x^{1/2} \mu_x}{e\kappa_{ph}}$$



L.D. Hicks and M.S. Dresselhaus,
Phys. Rev. B 47, 16631 (1993)

Thermoelectric Nanostructures: Results



Significant improvement over bulk materials

Mainly due to reduced lattice thermal conductivity

R. Venkatasubramanian et al., Nature 413 (2001) p. 597.

T.C. Harman et al., Science 297 (2002) p.2229

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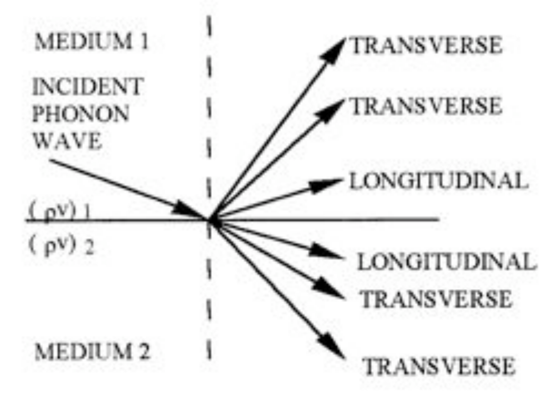
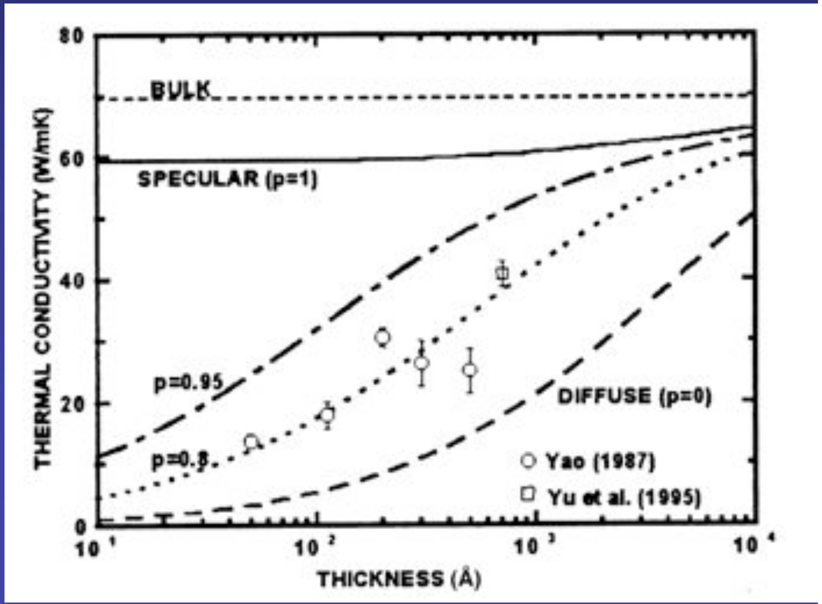
Phonons in Nanostructures

phonon transport



$$\kappa_{phonons} = \frac{1}{3}\int_0^{\omega_{max}} C(\omega)v(\omega)\Lambda(\omega)d\omega$$

$$\kappa_{phonons} = \frac{1}{4\pi}\int_0^{\omega_{max}}\left[\int_0^{2\pi}\sin^2\phi\, d\phi\int_0^{\pi} C(\omega)v(\omega,\theta,\phi)\Lambda(\omega,\theta,\phi)\cos^2\theta\sin\theta\, d\theta\right]d\omega$$



1. **Group velocity can be reduced in nanostructures**
2. **Anisotropic scattering in low dimensional systems**
3. **Limits of integrals**
4. **Heat capacity (small effect)**
5. **Diffuse scattering at interfaces**

Can we do better than diffuse scattering ?

G. Chen *, T. Zeng, T. Borca-Tasciuc, D. Song
Materials Science and Engineering A292 (2000)
155–161

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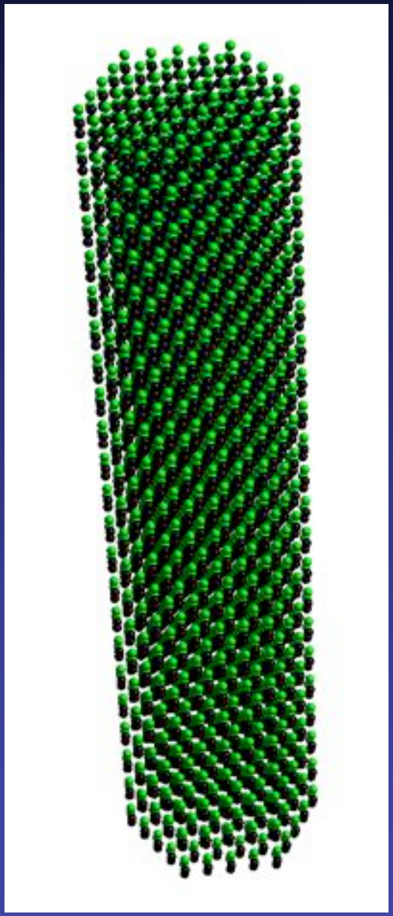
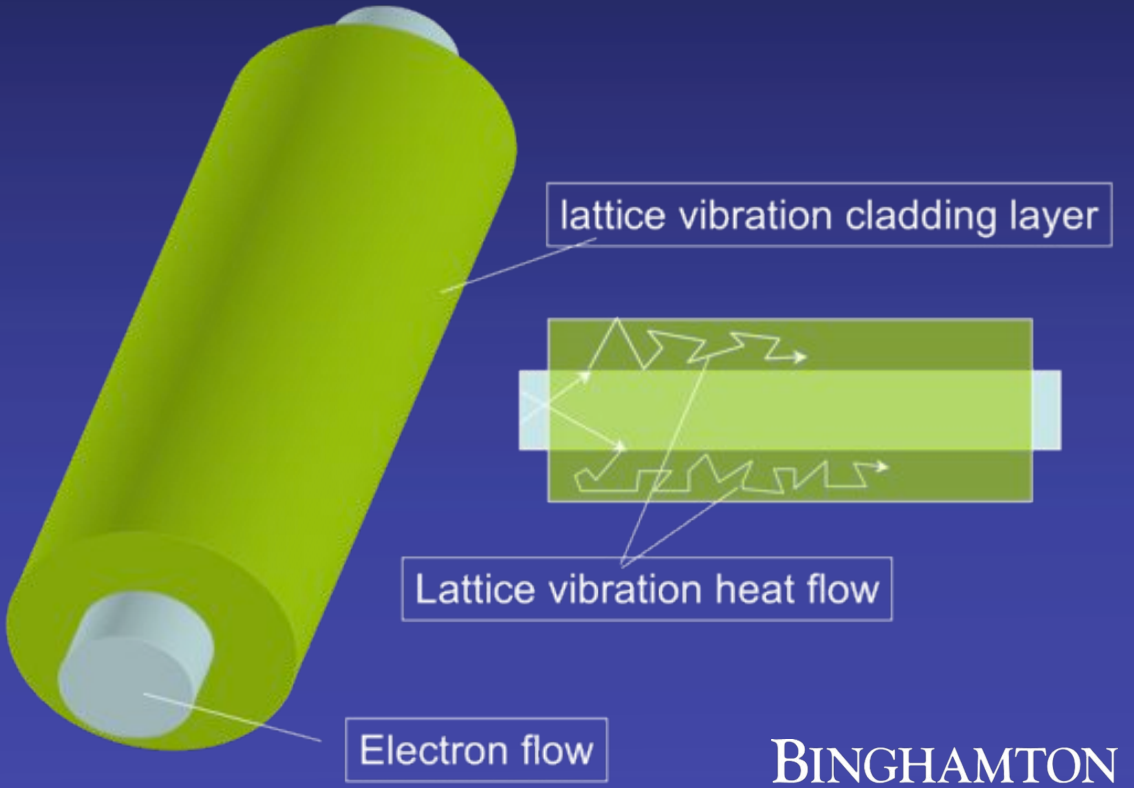
Low Energy Excitations at Thermoelectric Surfaces



Diffuse Scattering Limit: Phonon mean free path ~ nanowire diameter



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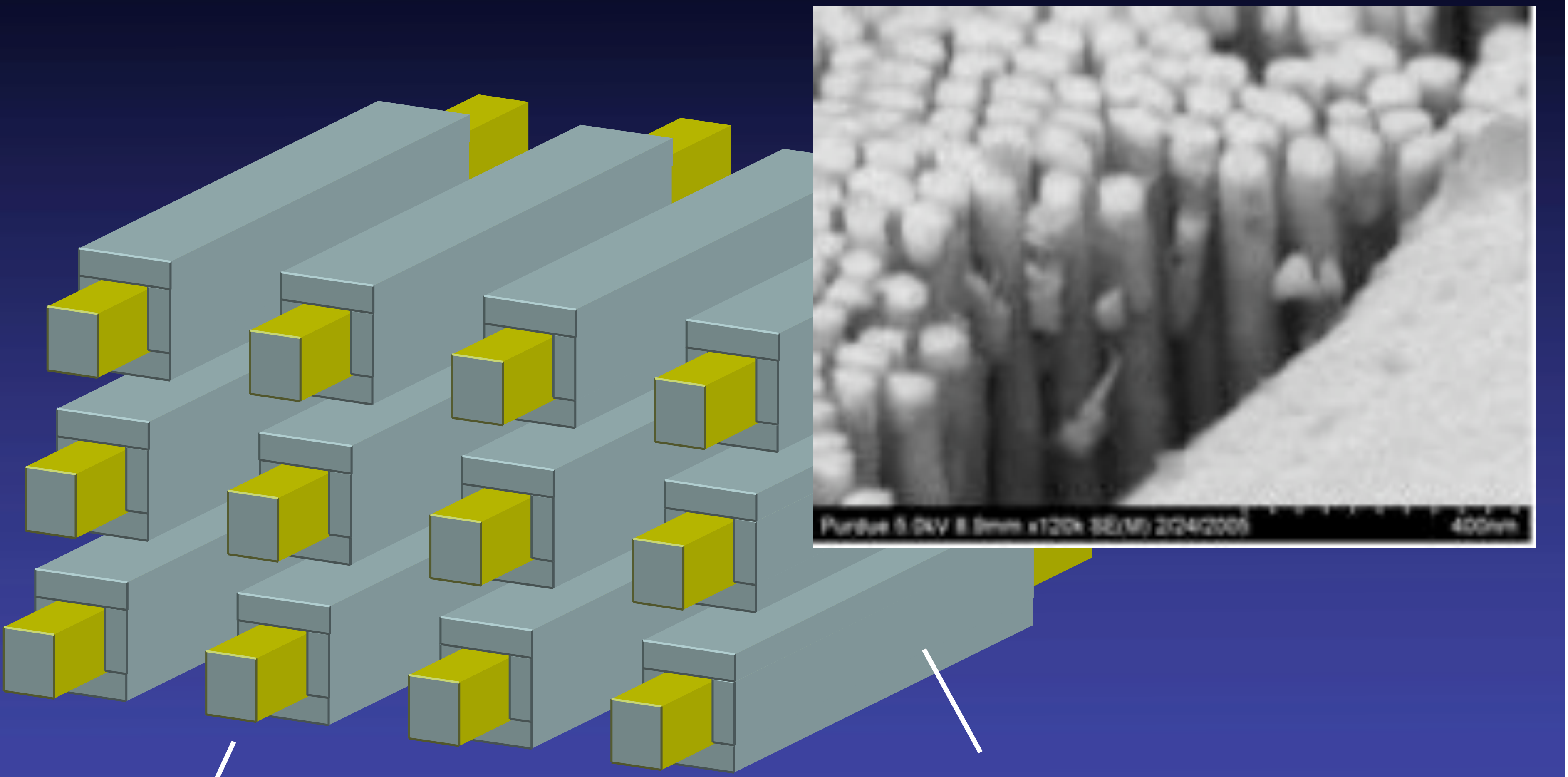
Nanocomposite Thermoelectrics?



Optimized electronic structure

Optimized phonon scattering structure

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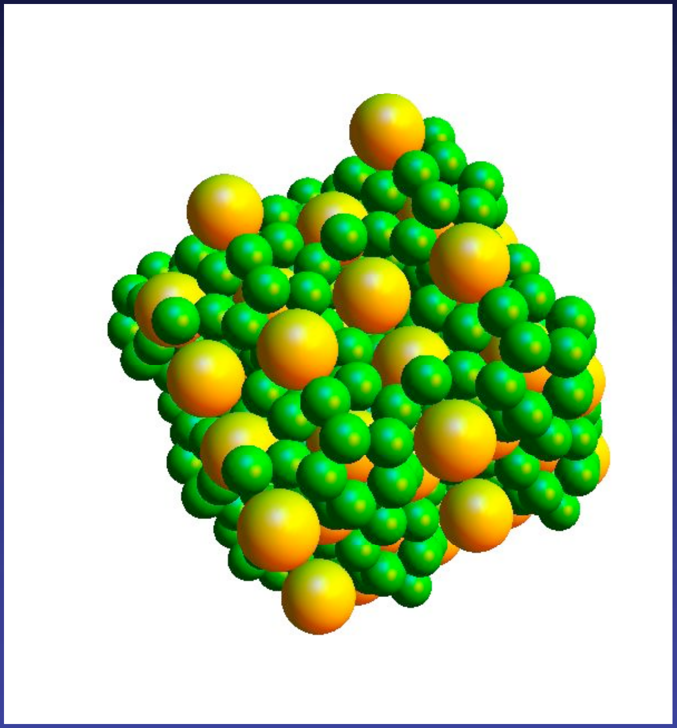
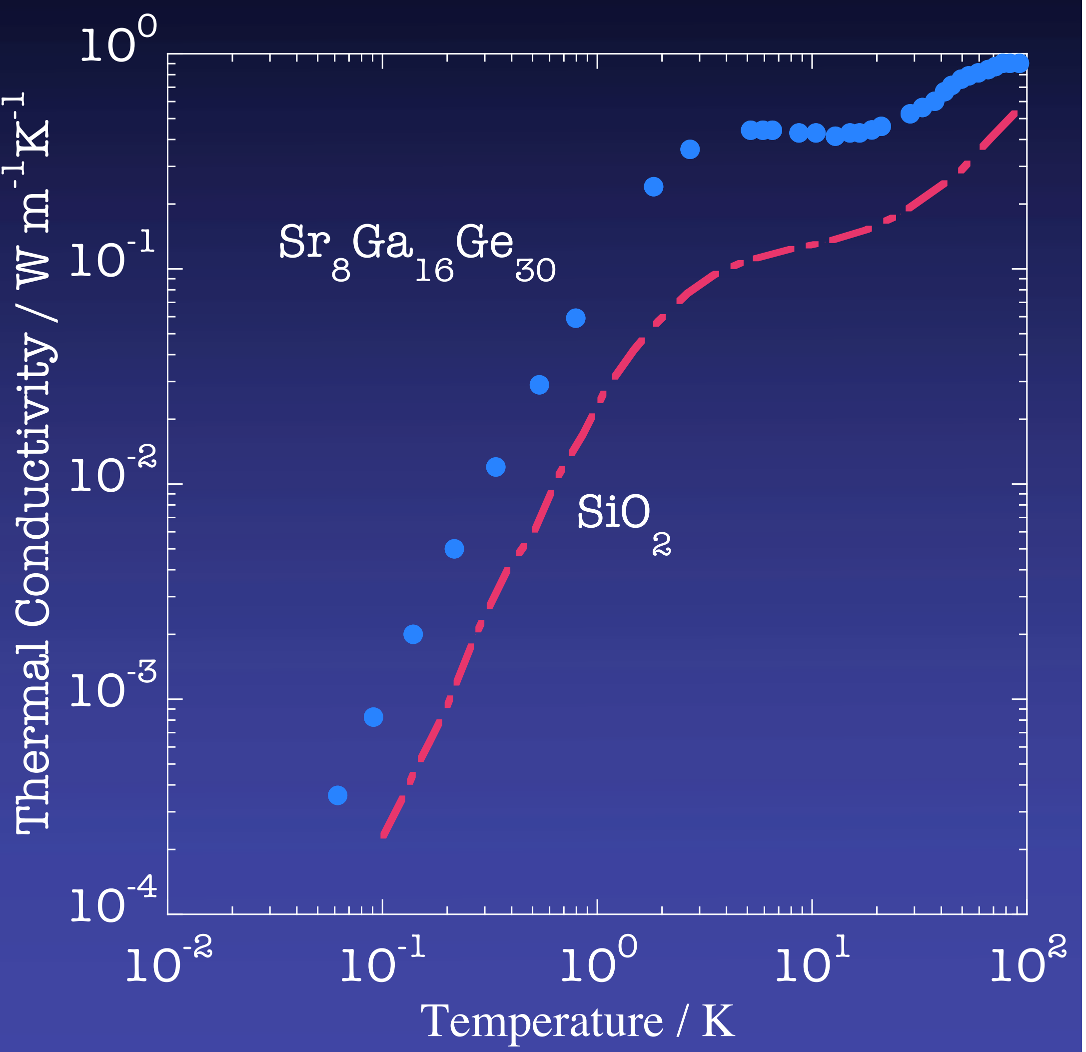
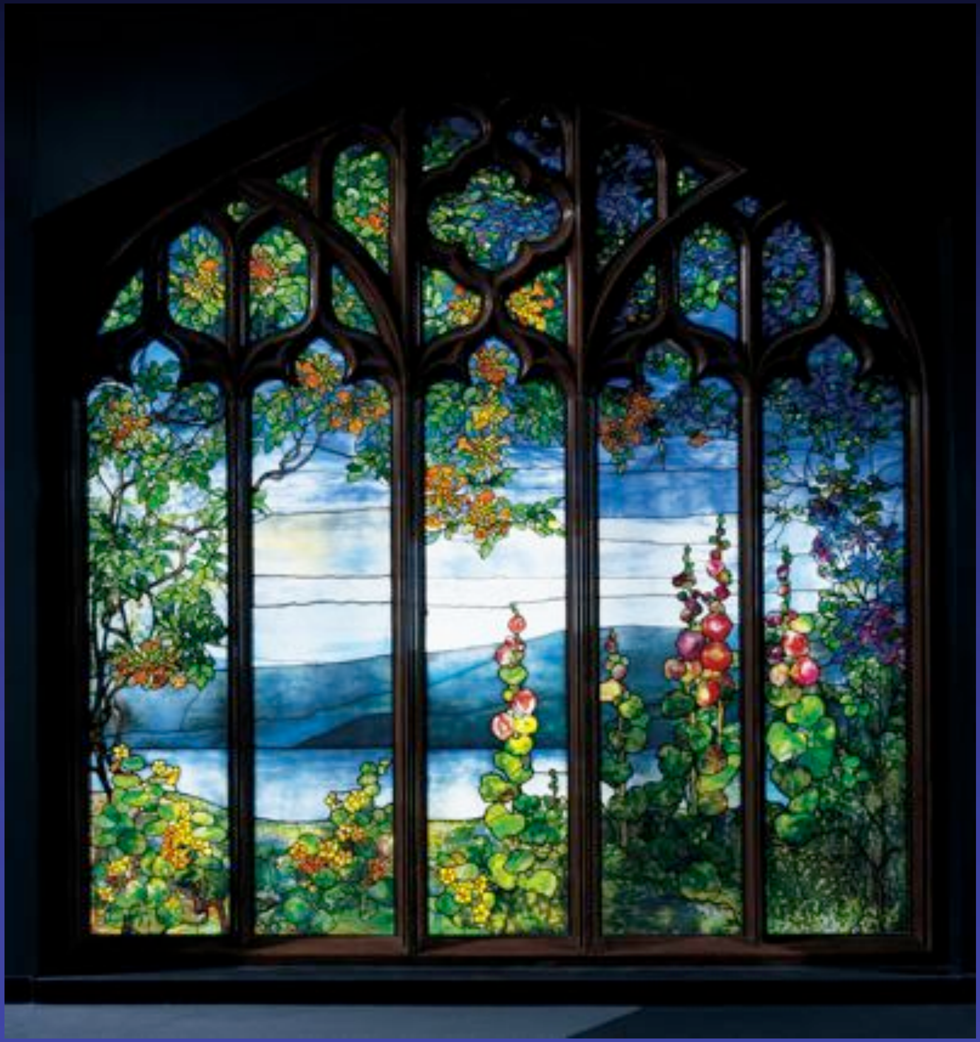
Electron Crystal - Phonon Glass





J. L. Cohn et al. Phys. Rev. Lett. 82, 779 (1999)
G.S. Nolas, D.T. Morelli, and Terry M. Tritt, Ann. Rev. Mat. Sci., 29, 89 (1999)

Transparent Thermoelectric Energy Generation?



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Conclusions

- Source of the universal and anomalous lattice vibrations in amorphous solids remains a mystery - Individual tunneling entities seem likely.
- TLS can be created in solids to produce phonon scattering (e.g. thin metal films).
- Ideas can be extended to nanoelectronic energy conversion devices to allow independent optimization of electronic and thermal properties.
- Are we close to creating artificial solids that mimic complex materials found in nature?