What makes glass interesting?

Caroline S. Gorham and David E. Laughlin
Department of Materials Science and Engineering Department
Carnegie Mellon University

Carnegie Mellon University

Abstract

Anomalous thermophysical properties of glasses have been studied for the past century; however glassy materials have often been overlooked for applications due to their incompatibility with periodic Bloch wave theorems. A recent wave of interest in glasses is driven by the quest for high thermo-power energy generation devices, since these devices operate most efficiently under glass-like thermal energy scattering conditions. Structural disorder in glasses results in the significantly reduced mean free paths of thermal energy transport compared with traditional phonons in crystalline counterparts. At the limit where the mean free paths are comparable to the excitation wavelengths, and the lifetimes compare with the excitation periods, confined vibrations drive the thermal properties. The character of elementary excitations and topological defects are the result of broken rotational symmetry through the super-cooled liquid to structural glass phase transition. An order parameter on the three-sphere, S^3, characterizes these intrinsic defects.

Thermo-physical property basics:

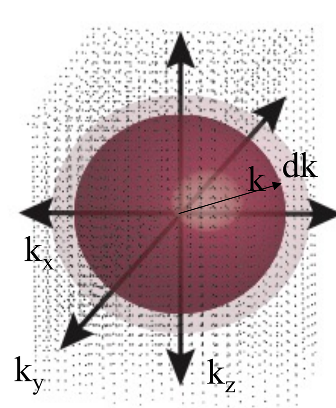
[Eigen]states, (ω_ι, e_i)

Frequencies are found by solving the eigenvalue equation,

$$\omega^2(\mathbf{k},\nu)\mathbf{e}(\mathbf{k},\nu) = \mathbf{D}(\mathbf{k})\mathbf{e}(\mathbf{k},\nu).$$

D: Dynamical matrix
ν: polarization
k: wavevector

Density of States, $g(k)dk$



The k-space volume of a single state per sphere in k-space:

$$g(k) = \frac{4\pi k^2}{\left(\frac{2\pi}{a}\right)^3} dk$$

The phase and group velocities:

$$v_p = \frac{\omega}{k} \quad v = \frac{d\omega}{dk}$$

Heat Capacity, C

Classical: $$C_V = \frac{dU}{dT}\Big|_{\delta W=0}$$

Quantum mechanical: $$C_V = \int \hbar\,\omega \frac{\partial f}{\partial T} g d\omega$$

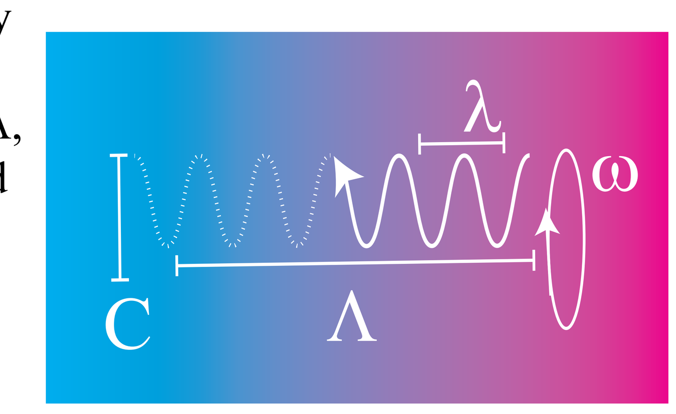
U: internal energy
δW: work done
f: Bose-Einstein distribution

$$f = \frac{1}{Ae^{\frac{\hbar\omega}{kT}} - 1}$$

Thermal Conductivity, κ

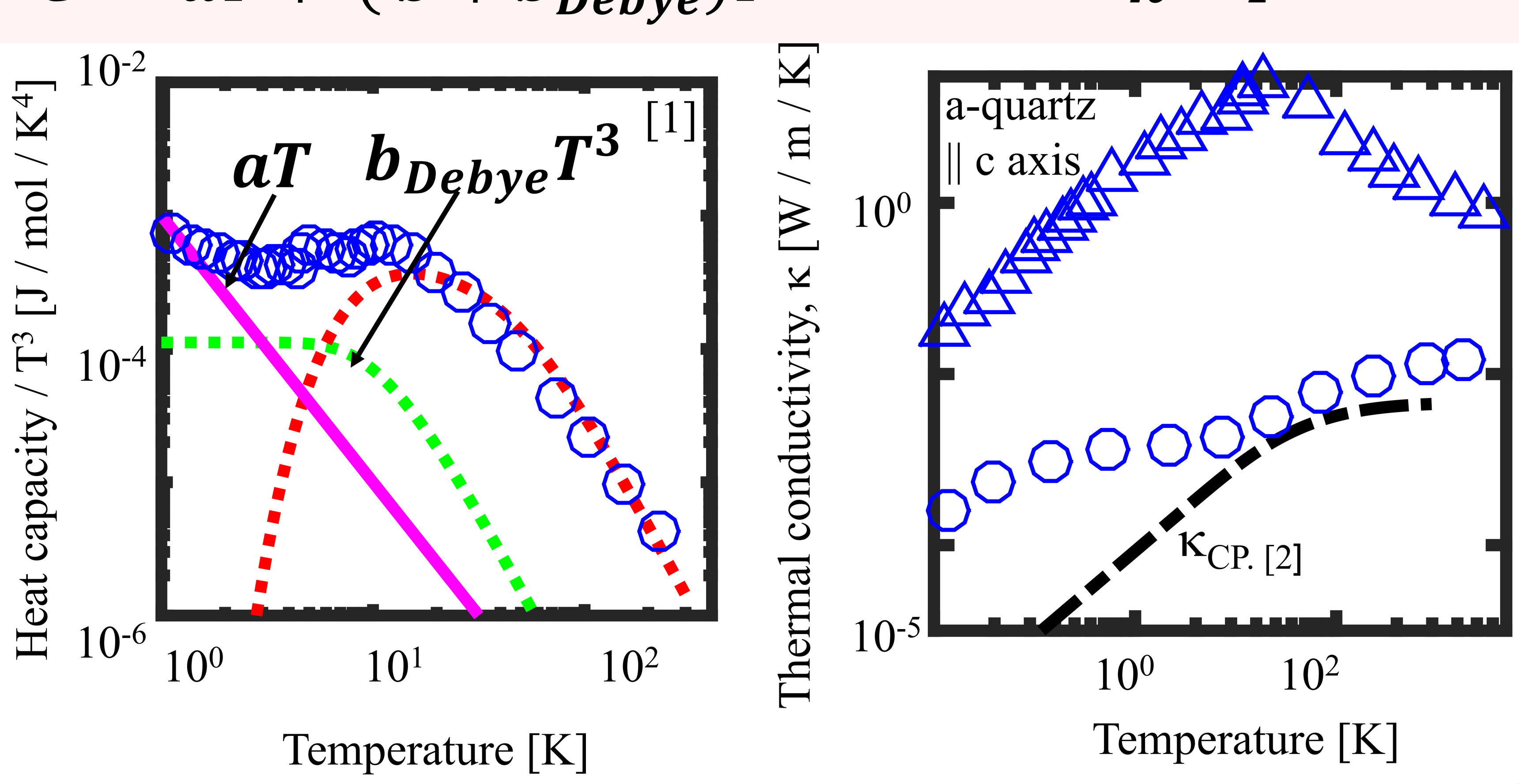
The thermal conductivity depends on how far thermal energy travels, Λ, how fast it travels, *v*, and how much energy it carries, *C*.

$$\kappa = \frac{Cv\Lambda}{3}$$

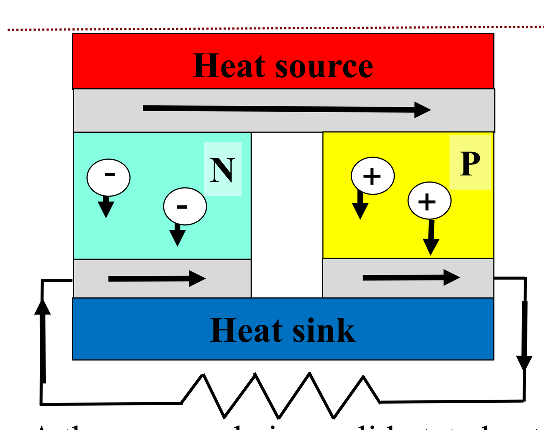


Ubiquitous thermal properties of glass:

$$C = aT + (b + b_{Debye})T^3 \qquad k \sim T$$



Thermoelectric device physics:

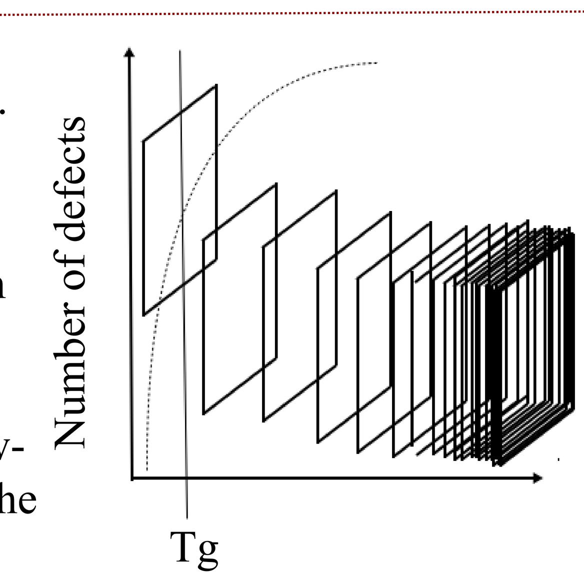


A thermocouple is a solid-state heat engine, i.e., with no moving parts, composed of materials with different thermopower coefficients.

A high dimensionless figure of merit, ZT, requires: low κ, high σ and high S.

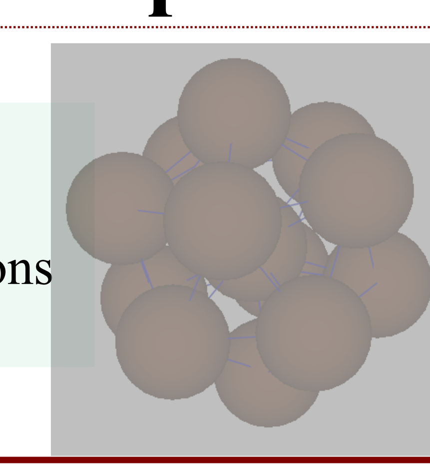
$$ZT = \frac{S^2\sigma T}{\kappa}$$

The magnitude and sign of S is driven by the entropy per charge carried by currents within the material. To characterize entropy per charge in low-κ systems, a detailed contribution of the defect density to entropy is necessary.



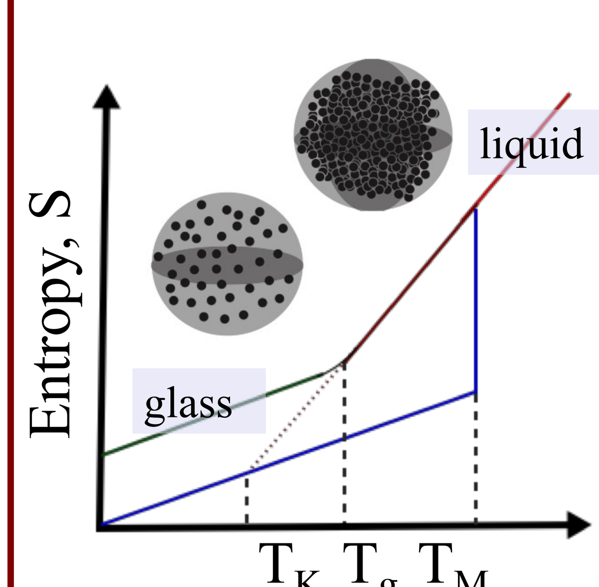
0. Characterize the phase transition:

1. Identify the broken symmetry
2. Define the order parameter
3. Examine the elementary excitations
4. Classify the topological defects



F. C. Frank found that in super-cooled liquids, topologically close-packing (TCP) results from the energetically favorable local arrangement of icosahedral clusters. [3]

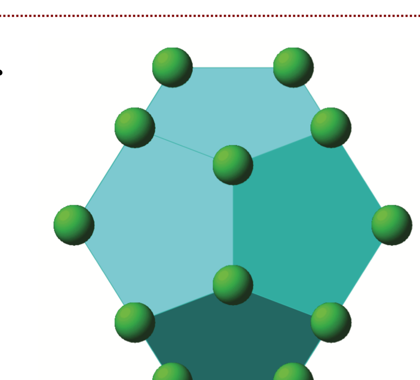
1. Identify the broken symmetry:



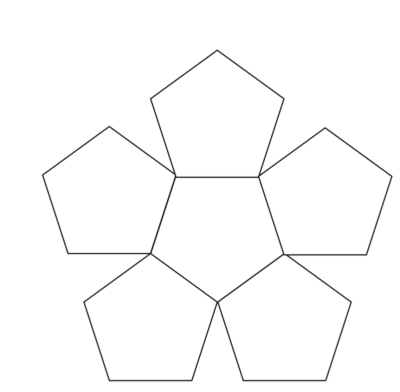
- A glass phase can only be formed through rapid cooling past the crystallization temperature, T_M.
- Anomalously low entropy of the supercooled liquid at the Kauzmann point, T_K, implies the existence of high order [4].
- The glass transition, T_g, is the result of high viscosity, as orientational degrees of freedom freeze.

2. Define the order parameter:

- An ideal glass is comprised of tetrahedral units that cover a sphere in four-dimensions. Intrinsic defects arise on packing Euclidean space.



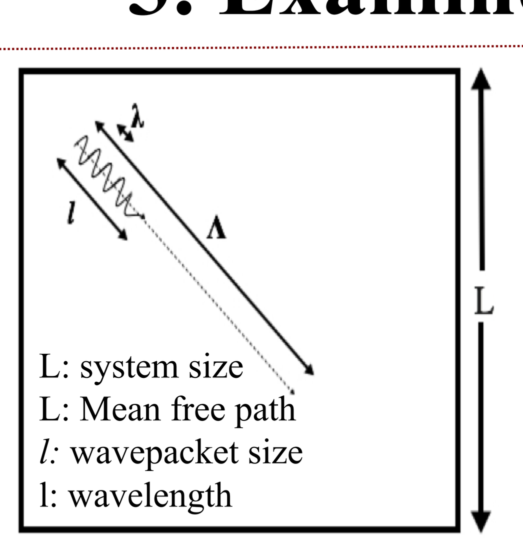
- As a lower-dimensional analogy, consider the defects created when unwrapping the covering of the two-sphere by pentagons into two-dimensions.



- The order parameter space for structural glass is the continuous group made by Hamilton's quaternion multiplication - the three-sphere, S^3

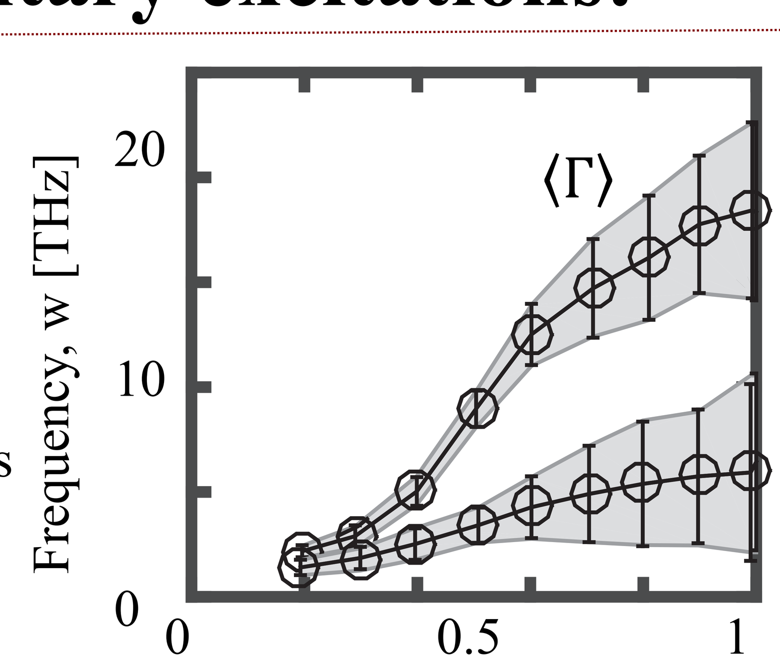
$$\mathbf{H} = \mathrm{a} + \mathrm{b}i + \mathrm{c}j + \mathrm{d}k\,; \quad i^2 = j^2 = k^2 = ijk = -1$$

3. Examine the elementary excitations:



Meaningful length scales in the phonon gas picture. For a phonon particle concept to be valid, λ << *l* << Λ << L.

The vibrational dispersion, ω(q), of amorphous silica exhibits widths, Γ, due to resonant broadening of structure factor peaks [5]. This is evidence of a radiative reaction. [6]



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Funding

C. S. G. is grateful for funding from NASA's Office of Graduate Research through the Space Technology Research Fellowship.