The influence of frozen-in stresses on the vibrational excitations in glasses

Walter Schirmacher

University of Mainz, Germany
Istituto Italiano di Technologia @Sapienza, Rome, Italy

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in collaboration with

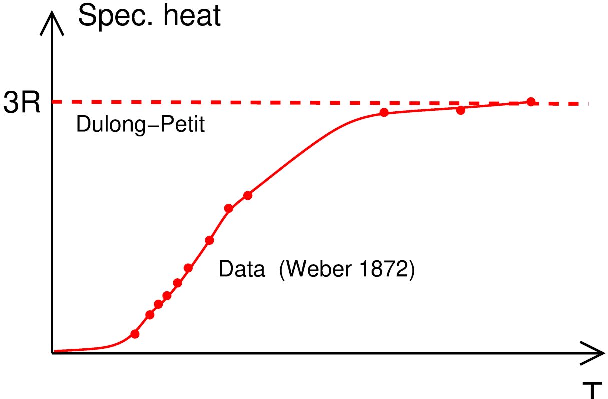
Giancarlo Ruocco, IIT and Sapienza Roma

Introduction: Specific heat of solids at low temperatures

Albert Einstein (1879 – 1955)

Peter Debye (1884 – 1966)

Introduction: Specific heat of solids at low temperatures



- At low temperatures the vibrational excitations (acoustic waves) obey quantum statistics

$$n(\omega) = \frac{1}{1 - e^{\hbar\omega/k_B T}}$$

$\omega = 2\pi f$

Specific heat and density of states

- The density of states $g(\omega)$ gives the frequency distribution of the vibrational excitations.
- The specific heat is related to $g(\omega)$ by

$$C(T) = k_B \int_0^\infty d\omega\, g(\omega) \left(\frac{\hbar\omega}{k_B T}\right)^2 \frac{e^{\hbar\omega/k_B T}}{(e^{\hbar\omega/k_B T} - 1)^2}$$

- Free acoustic waves: $g(\omega) \propto \omega^2$(Debye) $\Rightarrow$ $C(T) \propto T^3$ (Debye)
- In crystals this is experimetally observed for $T \to 0$.
- In glasses this is never observed!

Motivation of recent research: Low-frequency and low-temperature vibrational anomalies of glasses

- The low-frequency spectra of glasses and their associated low-temperature thermal properties remain poorly understood:
  - In very small computer-generated glasses, which do not allow for wave propagation, the DOS varies as
  $$g(\omega) \propto \omega^\gamma \text{ with } 2 \leq \gamma \leq 4\,;$$
  - The measured low-temperature specific heat does not obey Debye's $C(T) \sim T^3$ law, but varies as
  $$C(T) \propto T^{1+\delta} \text{ with } 0.2 \leq \delta \leq 0.7$$
  in the $T \sim 1\text{K}$ range;
- Better understood, but still controversal:
  - The DOS (1 THz range) and the specific heat (10 K range) exhibit a peak in $\dfrac{g(\omega)}{\omega^2} \quad \leftrightarrow \quad \dfrac{C(T)}{T^3}$ "boson peak"

Boson peak in $g(\omega)$ and $C(T)$

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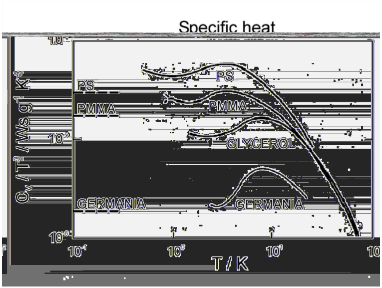
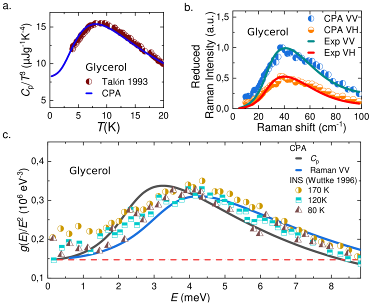


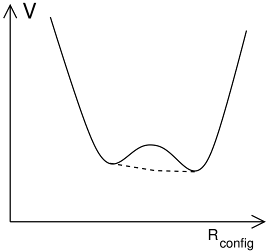


- This anomaly can be well described by spatially fluctuating elastic constants (Heterogeneous-elasticity CPA theory, Pan *et al.* 2021)
- Let's call the corresponding anomalous vibrations
  "type-I nonphononic vibrations´´

Traditional explanation of the low-T thermal anomalies in terms of double-well defects

- Tunneling model Anderson et al.(1972), Phillips (1972). It is assumed that quantum tunneling takes place between adjacent double-well potential minima. $\Rightarrow$ $C(T) \propto T$
- Soft-potential model Karpov et al. (1981). The energy landscape of the glass is supposed to exhibit potential minima with very small random second-order coefficients a_i and random local forces f_i

$$V(R) = \pm\frac{1}{2}a_i R^2 + \frac{1}{4!}bR^4 - f_i R$$



Double-well (soft) potential

- Resulting DOS: $\Delta g(\omega) \propto \omega^4$
- Resulting T dependence of the specific heat:

$$C(T) = AT + BT^3 + CT^5$$

- Caution: Here $g(\omega) \propto \omega^4$ (and hence $C(T) \propto T^5$) does not come from elastic disorder (Rayleigh scattering) but from an anharmonic

Generalized heterogeneous-elasticity theory allowing for frozen-in stresses

- Potential-energy density of (heterogeneous) elasticity theory:

$$\mathcal{V}_{\mathrm{harm}}(\mathbf{R}) = \sum_{\alpha\beta\gamma\delta} \varepsilon_{\alpha\beta}(\mathbf{R}) C_{\alpha\beta\gamma\delta}(\mathbf{R}) \varepsilon_{\gamma\delta}(\mathbf{R})$$

Here $C_{\alpha\beta\gamma\delta}(\mathbf{R})$ are the fluctuating elastic constants and $\epsilon_{\alpha\beta}(\mathbf{R}) = \frac{1}{2}[\partial_\alpha u_\beta + \partial_\beta u_\alpha]$.

- In disordered systems (glasses) an additional term arises:

$$\Delta\mathcal{V}_{\mathrm{harm}} = \sum_{\alpha\beta\gamma} \sigma_{\beta\gamma}(\mathbf{R}) \eta_{\alpha\beta}(\mathbf{R}) [\eta_{\alpha\gamma}(\mathbf{R}) - \varepsilon_{\alpha\gamma}(\mathbf{R})]$$

- In addition to the strains this term involves frozen-in stresses

$$\sigma_{\alpha\beta}(\mathbf{R}_{ij}) = \frac{1}{\Delta V} \frac{1}{r_{ij}} \phi'(r_{ij}) \mathbf{r}_{ij}^{\alpha} \mathbf{r}_{ij}^{\beta}\,.$$

and local vorticities

$$\eta_{\alpha\beta} = \frac{1}{2}\left[\partial_\alpha \mathbf{u}_\beta - \partial_\beta \mathbf{u}_\alpha\right].$$

- We call these stress-induced modes

"type-II" non-phononic excitations.

Generalized heterogeneous-elasticity theory

- Total Potential-Energy density:

$$\mathcal{V}_{\text{harm}} = \underbrace{\underbrace{\sum_{\alpha\beta\gamma\delta} \varepsilon_{\alpha\beta} C_{\alpha\beta\gamma\delta}(\mathbf{R})\varepsilon_{\gamma\delta}}_{HET} + \sum_{\alpha\beta\gamma} \sigma_{\beta\gamma}(\mathbf{R}_{ij})\eta_{\alpha\beta}[\eta_{\alpha\gamma} - \varepsilon_{\alpha\gamma}]}_{GHET}$$

- Setting the local stresses $\sigma_{\alpha\beta}(\mathbf{R}_{ij}) = 0$, one recovers Heterogeneus-elasticity theory, involving “type-I´´ excitations (boson peak)

- On the other hand, assuming $C_{\alpha\beta\gamma\delta} \equiv$ const., but keeping the $\sigma_{\alpha\beta}(\mathbf{R}_{ij})$ finite. gives a theory for the influence of local stresses on the low-frequency vibrational spectrum of glasses[4]
and their low-T thermal anomalies[5].

[4] WS, et al. Nature Comm. 15, 3107 (2024)

[5] WS, G. Ruocco, Phys. Rev. Lett. 135, 126102 (2025)

Type-II nonphononic excitations: Fluctuating local stresses
1. DOS of Computer-generated very small samples

– The DOS is calculated via the eigenvalue spectrum $\rho(\lambda)$ according to

$$g(\omega) = 2\omega\rho(\lambda = \omega^2)\,.$$

- The spectrum (and therefore the DOS) is intimated related to the distribution of small stresses $P(\sigma)$;
- The stress-induced low-frequency spectrum has a (small)direct contribution and a (dominant) indirect one, mediated by the type-I states:

$$\rho_{\text{dir}}(\lambda) \sim P(\sigma)\Big|_{\lambda\sim\sigma} \qquad \rho_{\text{indir}}(\lambda) \sim \sigma^2 P(\sigma)\Big|_{\lambda\sim\sigma}$$

- In computer simulations one standardly uses potentials with smooth cutoff
- The smoothing (tapering) guarantees that the mth derivative is continuous at the cutoff $r = r_c$, i.e.

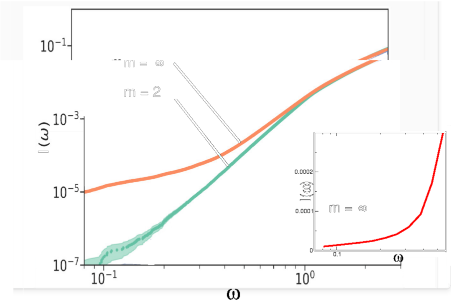
$$\phi(r \approx r_c) \sim (r_c - r)^{m+1} \quad \Rightarrow \quad P(\sigma) \sim \sigma^{\frac{1}{m}-1}$$

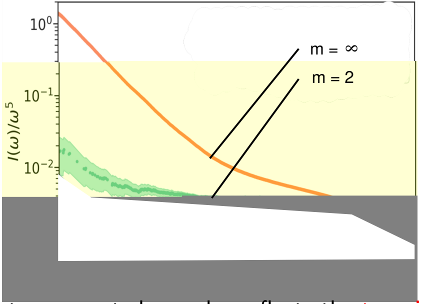
Type-II nonphononic excitations: Fluctuating local stresses

1. DOS of Computer-generated very small samples

$$\phi(r \approx r_c) \sim (r_c - r)^{m+1} \quad \Rightarrow \quad g_{\substack{\mathrm{dir}\\ \mathrm{indir}}}(\omega) \sim \begin{cases} \omega^{\frac{2}{m}-1} = \omega^0 & \text{for } m=2 \\ \omega^{\frac{2}{m}+3} = \omega^4 & \text{for } m=2 \end{cases}$$

$$I_{\mathrm{dir}}(\omega) = \int_0^{\omega} d\bar{\omega} g(\bar{\omega}) \sim \begin{cases} \omega & m=2 \\ \ln\omega & m=\infty \end{cases} \qquad I_{\mathrm{indir}}(\omega) = \int_0^{\omega} d\bar{\omega} g(\bar{\omega}) \sim \begin{cases} \omega^5 & m=2 \\ \omega^4 & m=\infty \end{cases}$$





- The low-frequency DOS of small computer-generated samples reflects the tapering and is not any generic feature of a glass.

Type-II nonphononic excitations: Fluctuating local stresses
2. Specific heat of macroscopic samples

- In macroscopic samples the Debye waves are present, which are absent in the microscopic computer glasses. This modifies the DOS:
  - The hybridization with the waves modifies the type-II DOS;
  - Below the BP the Debye DOS $g_D(\omega) \sim \omega^2$ is present.

  Taking only transverse waves into account, we get $(\lambda = \omega^2)$

$$\rho_{\text{dir}}(\lambda) \sim P(\sigma)\Big|_{\lambda=f(\sigma)} \qquad \rho_{\text{indir}}(\lambda) \sim \sigma^2 P(\sigma)\Big|_{\lambda=f(\sigma)}$$

  with

$$f(\sigma) = \frac{1}{\zeta}\sigma\left(1 + \frac{\sigma}{G}\right), \quad \zeta = \frac{1}{2}\rho_m\langle r_\perp^2\rangle = \text{inertia density}$$

from hybridization with the waves ↑ $\qquad \sqrt{\langle r_\perp^2\rangle}$=gyration radius of vortices

- Generic interatomic potential near its minimum value

$$\phi(r{\approx}r_{\text{min}}) \sim (r - r_{\text{min}})^2 \quad \Rightarrow \quad P(\sigma) = \text{const.}$$

Type-II nonphononic excitations: Fluctuating local stresses
2. Specific heat of macroscopic samples

- Resulting DOS

$$g(\omega) = 2\omega\rho(\omega^2) = g_D(\omega) + g_{\text{dir}}(\omega) + g_{\text{indir}}(\omega)$$

$$\underbrace{g_{\text{dir}}(\omega) \sim \omega \frac{1}{\sqrt{1+\frac{\omega^2}{\omega_0^2}}} \approx \omega^r}_{\rightarrow \text{ low}-\text{T specific heat}} \qquad 0<r<1 \qquad \underbrace{g_{\text{indir}}(\omega) \sim \omega^5 \frac{1}{\sqrt{1+\frac{\omega^2}{\omega_0^2}}} \approx \omega^{4+r}}_{\rightarrow \text{ modification of the boson peak}}$$

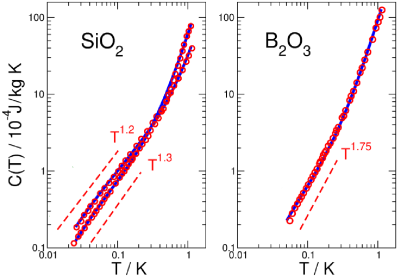
$$\omega_0^2 = \frac{G}{2\zeta} = \frac{V_T^2}{\langle r_\perp^2 \rangle}$$

Contribution from
hybridization with the waves

- Specific heat $C(T)$

$$\sim \int_0^\infty d\omega(\beta\omega)^2 \frac{e^{\beta\omega}}{[e^{\beta\omega}-1]^2}[g_D(\omega) + g_{\text{dir}}(\omega)]$$

$$\sim T^{1+r} \text{ near } 0.1 \text{ K}$$



Type-II states and "quasilocalized" states

- The term quasilocalized states has been coined by Schober and Oligschleger (1996), who found localized low-frequency non-phononic vibrational excitations in very-small-sample simulations.
- They argued that in larger samples, which allow for phonons, such states should hybridize with the phonons and become delocalized.
- Such non-phononic vibrational states were supposed to exist in addition to the elastic waves (phonons).
- Later the quasilocalized states were associated with defect states due to anharmonic soft-potential configurations, which, according to the model of Karpov et al. (1981) exhibit a DOS $\propto \omega^4$.
- Numerical evidence for the existence of defect states with an ω^4 DOS Lerner, Bouchbinder (2021) is questioned by the possibility that such a behavior may be an artifact of the tapering.
- We argue that indeed quasi-localized states exist. They are the type-II states, associated with frozen-in stresses.

Conclusions

- In the presence of local frozen-in stresses local vibrations appear, which form vortices ("Type-II nonphononic states").
- The Type-II spectrum is related to the distribution of the local stresses.
- In very small computer-generated systems, where low-frequency waves do not exist, the observed $g(\omega) \sim \omega^s$ behavior of the spectra may be related to the smooth cutoff of the potential (tapering).
- In macroscopic samples the type-II excitations hybridize with the Debye waves and may be classified as quasi-localized states in the sense of Schober and Oligschleger.
- These states may be responsible for the anomalous low-T behaviour of the specific heat, instead of quantum tunneling of bi-stable configurations.