The "Tunnelling Two-Level Systems" Model of the Low-Temperature Properties of Glasses: Successes, Problems, Prospects*

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cf: AJL and D.C. Vural, J. Phys. Chem. B 117, 12966 (2013)

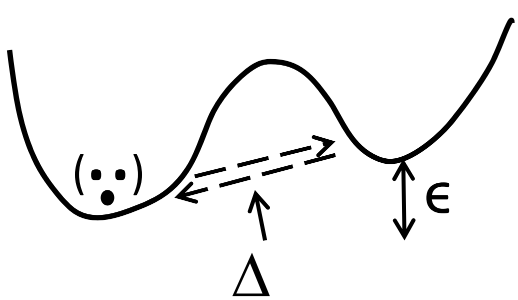
ILLINOIS

SOME PROPERTIES OF SOLIDS BELOW 1K

	Crystals	Glasses
Specific heat	$\sim T^3$	$\sim T$
Thermal conductivity	$\sim T^2$ ($\times \exp -a\theta_D/T$)	$\sim T^2$
Ultrasonic absorption	$\sim \omega^4$	$\sim \omega^2/T$ (for $\omega \ll T$)
Hysteresis?	no	yes
……………………..	……..	…….

The (tunnelling) two-level systems (TTLS) model
(Anderson, Halperin + Varma 1972, Phillips 1972):

Intuitively:



$$\hat{H}_{TLS} = \frac{1}{2}\begin{pmatrix} \epsilon & \Delta \\ \Delta & -\epsilon \end{pmatrix}$$

Precisely:

a) "TLS" model:

$$\hat{H} = \hat{H}_{ph} + \hat{H}_{TLS} + \hat{H}_{int}$$

$$\hat{H}_{ph} = \sum_{\boldsymbol{k}\alpha} \hbar\omega_{k\alpha} a^{+}_{k\alpha} a_{k\alpha}, \quad \omega_{kd} = c_{\alpha}|k| \quad \left[a_{k\alpha}, a^{+}_{k'\beta}\right] = \delta_{kk'}\delta_{\alpha\beta}$$

i.e "ordinary" phonons

$$\hat{H}_{TLS} = \sum_{i} E_i b_i^{+} b_i \qquad \{b_i, b_i^{+}\} = 1, \qquad \left[b_i, b_j^{+}\right] = 0 \text{ for } i \neq j$$

i.e. Pauli operators

$$\hat{H}_{int} = \sum_{\alpha\beta} \int \underset{\substack{\uparrow \\ \text{phonon strain}}}{\hat{e}_{\alpha\beta}} \hat{T}_{\alpha\beta}(\boldsymbol{r}) d\boldsymbol{r} \qquad \hat{T}_{\alpha\beta}(r) \equiv \sum_{i} g^{(i)}_{\alpha\beta\gamma} \hat{\sigma}^{i}_{\gamma} \delta(\boldsymbol{r} - \boldsymbol{r_i})$$

“tunnelling”

b) <u>“TTLS” model imposes further constraints</u>:

$$\hat{H}_i = \frac{1}{2}\begin{pmatrix} \epsilon_i & \Delta_\mathrm{i} \\ \Delta_i & -\epsilon_i \end{pmatrix} \qquad \left(\epsilon_i \equiv \sqrt{\epsilon_i^2 + \Delta_i^2}\right)$$

$$\rho(\epsilon, \Delta) = \text{const.}/\Delta \Rightarrow \rho(E) = \text{const.}\ (\equiv \bar{P}_0)$$

$$g_{\alpha\beta\gamma}^{(i)} = \frac{g_{\alpha\beta}^{(i)}}{E_i}\left(\epsilon_i \delta_{z\gamma} + \Delta_i \delta_{x\gamma}\right), \left(\sigma_z \equiv \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}, \sigma_x \equiv \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}\right)$$

with $g_{\alpha\beta\gamma}^{(i)}$ having random (e.g. Gaussian) distribution with rms value $\bar{g}$.

(may be different for L and T phonons)

Note with this form $g^{(i)}_{\alpha\beta\gamma}$ is strongly peaked towards small values

In this talk, I will define ("weakly interacting") TTLS model by the above assumptions plus the assumption that the correct explanation of any given physical property is given by a calculation to the lowest order in $\bar{g}$ which gives a nontrivial result (e.g. 0^{th} for C_v, 1^{st} for US absorption and κ …)

Some successes of the TTLS model (as so defined):

predicts $C_v(T) \propto T$ (✓) (actually $T^{1+\alpha}$, $\alpha \sim 0 \cdot 1 - 0 \cdot 3$)
`` $\kappa(T) \propto T^2$ (✓) (actually $T^{2-\beta}$, $\beta \sim 0 \cdot 1 - 0 \cdot 3$)
`` $\alpha(\omega, T) \propto \omega \tanh \omega/2T$ ✓
`` saturation, echoes … ✓
`` log'c dependence of $C_v(t)$ ✓

Moreover, in some amorphous systems (e.g. polyethylene) fairly direct evidence (e.g. from luminescence of embedded organic molecules) for TLS (↑: at room temp. not (directly) at ≤ 1K). Also oxide Josephson junctions, KBr – KCN …

Prediction very specific to TTLS assumption (Jäckle, 1972): in both high-ω, low T (“resonance”) and low- ω, high-T (“relaxation”) regimes, Q-factor for (linear) ultrasound absorption is constant:

$$Q_{res}^{-1} = \pi C$$

$$C \equiv \overline{P_0}\gamma^2/pc^2$$

$$Q_{rel}^{-1} = \frac{\pi}{2} C$$

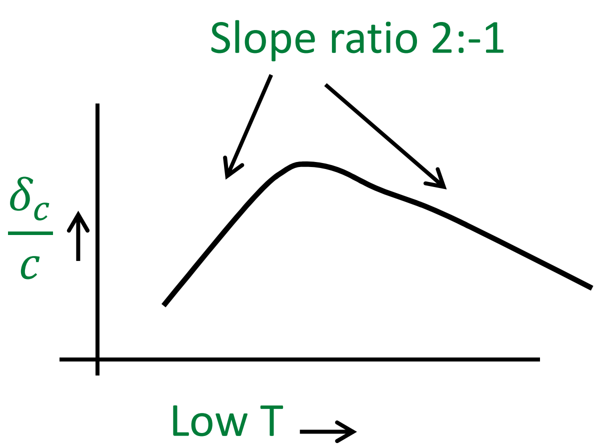
Direct measurement of Q^{-1} subject to considered WWWW, but can relate by KK to velocity shift: since Q_{res}^{-1} has low-frequency. ω ∝ T cutoff Q_{rel}^{-1} has high-frequency $\omega \propto T$ cutoff , we get (up to additive constants)

$$\left.\frac{\delta c}{c}\right|_{res} = C \ln\left(\frac{T}{T_0}\right)$$

$$\left.\frac{\delta c}{c}\right|_{rel} = -\frac{C}{2} \ln\left(\frac{T}{T_0}\right)$$

Thus:

prediction:



This general pattern is indeed seen. But …

Some problems with the TTLS scenario

1. Except in a few very special cases (KBr-KCN, Al_2O_3 JJ's …) no clear picture of the nature of the TTLS.

2. Does not by itself explain drastic change in experimental properties of glasses above 1K (eg plateau ~ 1- 10 K in thermal cond^{y}.)

3. At least one specific prediction definitely wrong (at least in SiO_2, Bk7): in plot of $\delta c/c$ vs ln T, which general shape right, slope ratio is not 2:-1 but 1:-1. No simple modification of TTLS postulates appears able to fix this.

4. Universality of Q^{-1} (measurable by velocity shift and use of KK relation). In TTLS model,

$$Q_{res}^{-1} = \pi C. \qquad C = \frac{\overline{P_0}\gamma^2}{\rho c^2}$$

In C, 4 factors, each fluctuating between materials by factor ~ 5-10; no verticles nevertheless for ~ 30 different materials

$$Q_{res}^{-1} = 3 \times 10^{-4} \qquad \pm \sim 50\%$$

Is TTLS model successful because it is unique, or because it is a special case of a much more general scenario?

“crystals are the anomaly, glasses the norm!”

Alternative “collective” scenario (CCYu and AJL 1988, Burin & Kagan 1996, DC Vural and AJL 2011):

whatever non-phonon excitations we start with (maybe TTLS?) dominant effect phonon-mediated interaction.

The (generalized) collective scenario:

$$\hat{H} = \hat{H}_{ph} + \hat{H}_{np} + \hat{H}_{int}$$

$\hat{H}_{np}$ specified by (possibly random) matrix elements.
Quantity of fundamental interest is (non-phononic) stress tensor

$$\hat{T}_{ij} \equiv \frac{\partial \hat{H}_{np}}{\partial e_{ij}}$$

$$\Rightarrow H_{int} = \sum_{ij} \int d\boldsymbol{r}\, \hat{e}_{ij}(\boldsymbol{r}) \hat{T}_{ij}(\boldsymbol{r})$$

Elimination of phonons leads to effective stress-stress interaction (Joffin & Levelut 1975):

$$H_{np}^{(eff)} = \sum_{ijkl} \int d\boldsymbol{r} \int dr \frac{\Lambda_{ijkl}(\hat{n}_{rr'})}{|r - r'|^3} \hat{T}_{ij}(\boldsymbol{r}) T_{kl}(\boldsymbol{r}')$$

$\Lambda_{ijkl}(\hat{n}_{rr'})$ = nasty 4th- rank tensor

Conjecture: $H_{np}^{(eff)}$ dominates over orginal $\hat{H}_{np}$

In view of $|\boldsymbol{r}-\boldsymbol{r}'|^3$ dependence, problem is self-similar ⇒ expect real-space normalization procedure to scale to fixed point.

DC Vural & AJL 2011 (cf. Burin & Kagan 1996): universal value of Q^{-1} due to fact that absorbed entity (phonon) identical to one whose exchange generates effective interaction. Small value of Q^{-1} a result of (a) multiplicity of phonon modes and stress-tensor matrix elements (b) logarithmic factor arising from real-space scaling (indeed, predict that as $L \to \infty$,

$$Q^{-1} \sim \left(\ln\left(\frac{L}{L_0}\right)\right)^{-1/2} \to 0!)$$

Obvious question: similar effects in electrodynamics of complex media? (note: in many glasses such as SiO_2, electric-dipole interactions may be comparable to stress-stress.)

<u>“Smoking – gun” tests?</u>

Problem: alternative scenario at present too generic to make many specific predictions. So as Aunt Sally, choose scenario as different as possible from TTLS while not simply SHO:

$$\hat{T}_{ij} \text{ is random matrix}$$

1. Temperature dependence of ultrasound absorption:

$$\left.\begin{aligned} Q^{-1}_{TTLS}(\omega, T) &= \text{const.}\, x \tanh(\hbar\omega/2k_B T) \\ Q^{-1}_{RM}(\omega, T) &= \text{const.}\, x \tanh(1 - \exp - \hbar\omega/k_B T) \end{aligned}\right\} \text{distinguishable!}$$

2. Low-T properties of amorphous toluene:

   fluorescence of organic molecules embedded in eg PET (typical “glass”) seems to reflect TLS characteristics. However, similar experiments on solid amorphous toluene give <u>no</u> evidence for TLS. Thus, if we can measure $T < 1\text{K}$ properties of solid amorphous toluene:

   if very different from typical glass, supports TLS hypothesis

   if similar to other glasses, suggests TLS model is <u>not</u> the explanation.

Happy birthdays,
Phil and Freeman!