

Università dell'Insubria, Como

*Anomalous Multilevel Tunneling
Systems in Cold Multicomponent
Amorphous Solids*

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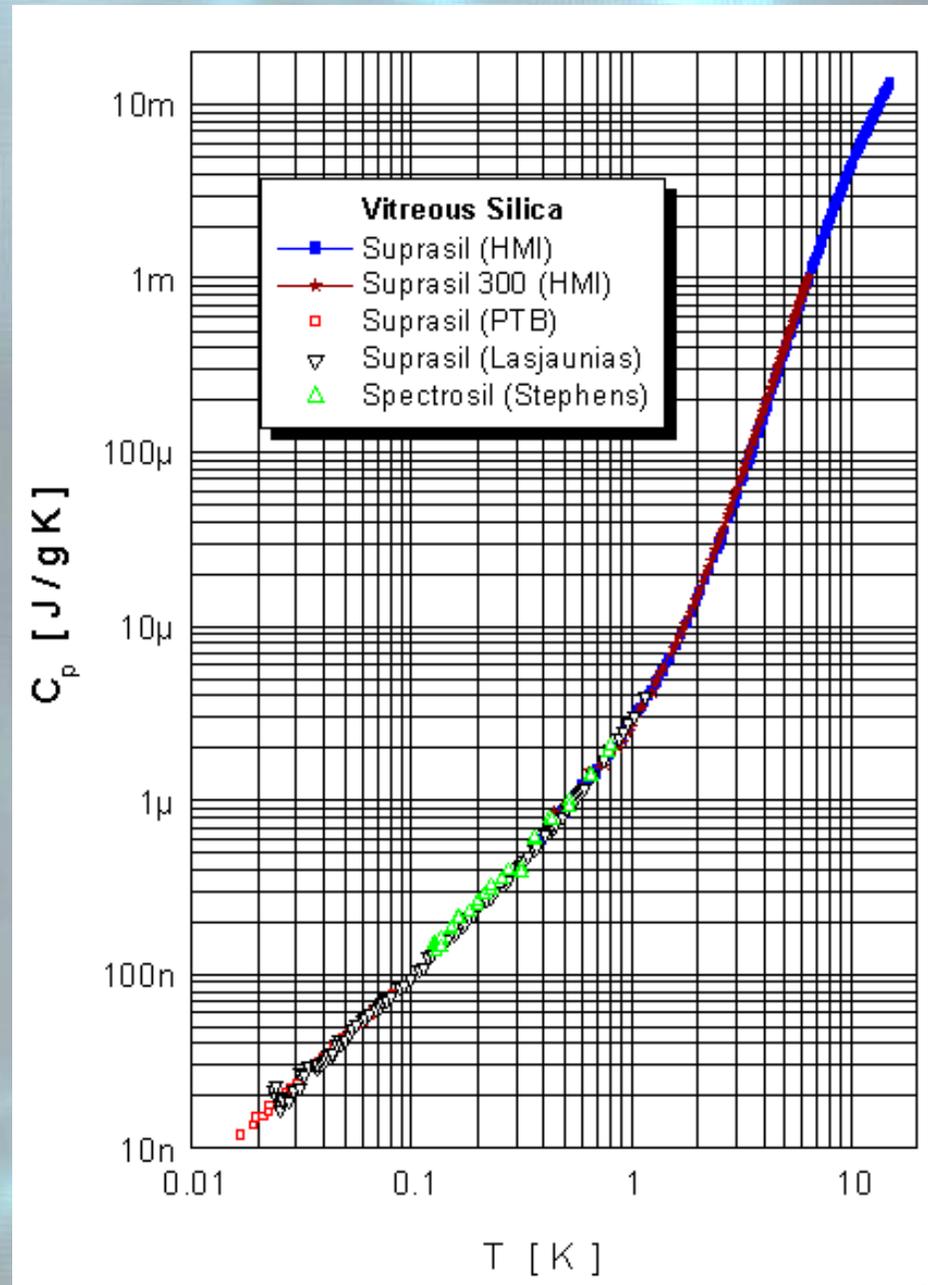
Plan of Talk:

- **Physics of cold glasses and standard theory**
- **New effects in multicomponent glasses**
- **Present explanation (Two-Network Model) & results (Permittivity and Specific Heat)**
- **Role of multilevel tunneling systems at $B=0$ (Evidence from mixed glasses)**
- **Conclusions**

PHYSICS OF COLD GLASSES

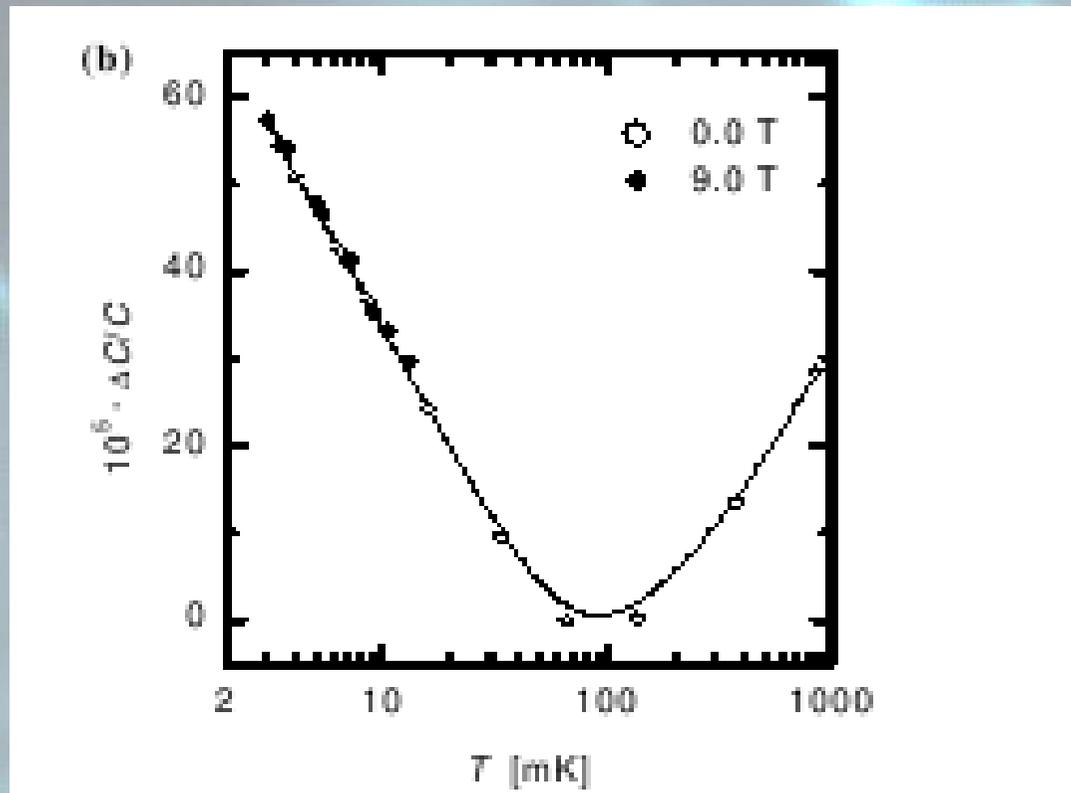
- Glasses at low temperatures (<1 K) show UNIVERSAL physical features (in heat capacity, dielectric constant and sound velocity)
- To be used as **sensors for low-T thermometry**
- Maybe as sensors for low-T magnetometry ?

Universal $T+T^3$
heat capacity
temperature
dependence
(2LS+phonons)
of **pure** glass



Universal $\ln(T)$ temperature dependence of electrical capacitance (note: no change in $B=9$ T)

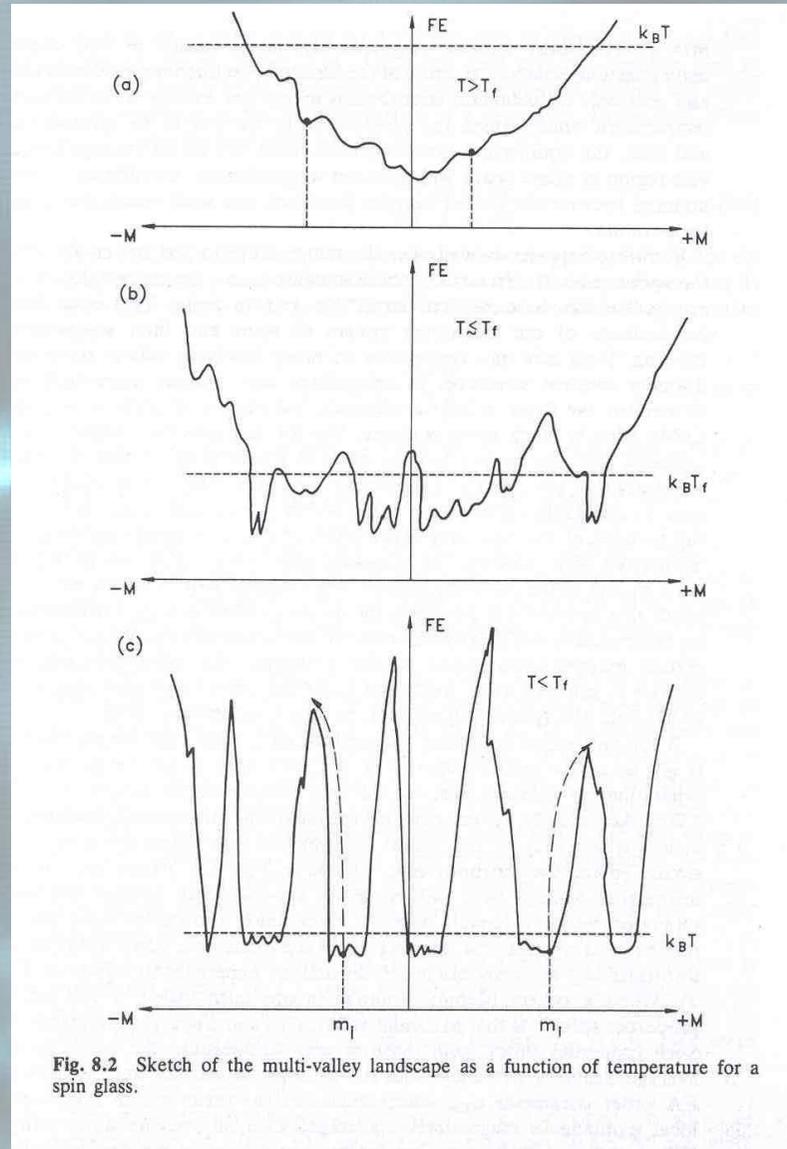
- pure a-SiO₂



Theoretical Interpretation: 2-level Systems

- Glasses are characterised by **many minima** in some configuration-space **energy landscape**
- At low temperatures, only **very few and very deep minima** are available for excitations => **double**-well potentials
- 2-level system tunneling model explains a number of experimentally observed features (Anderson, Halperin & Varma 70s)

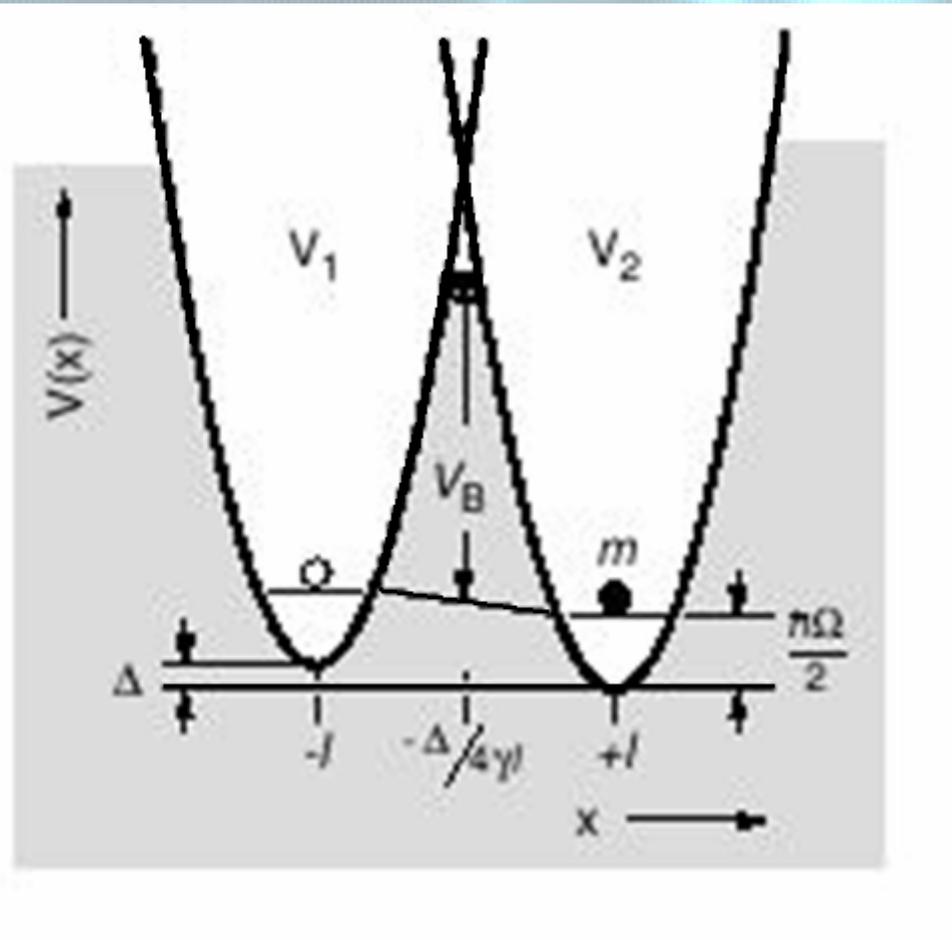
Energy landscape of a glass



Two-level Systems:

$$\Delta = E_1^{(0)} - E_2^{(0)}$$

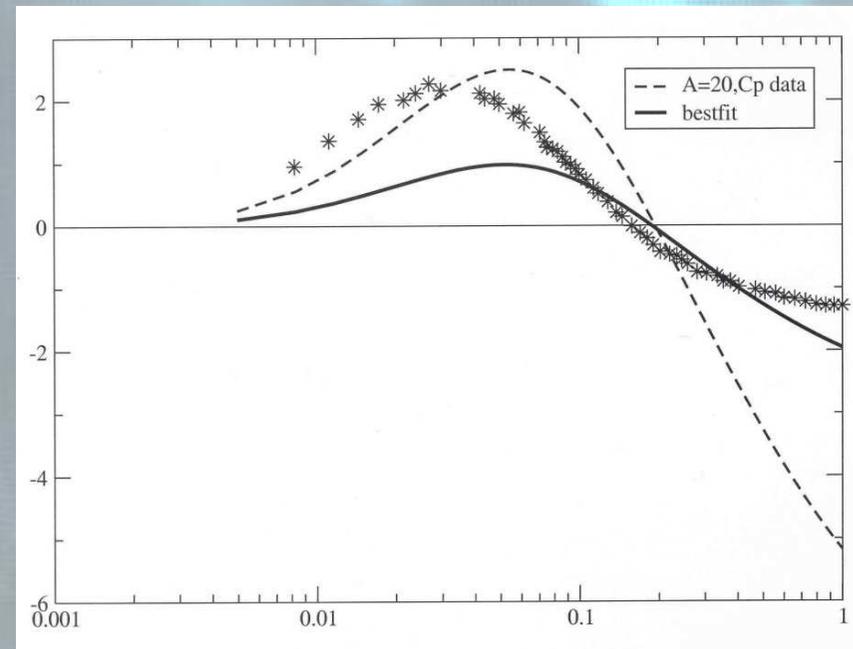
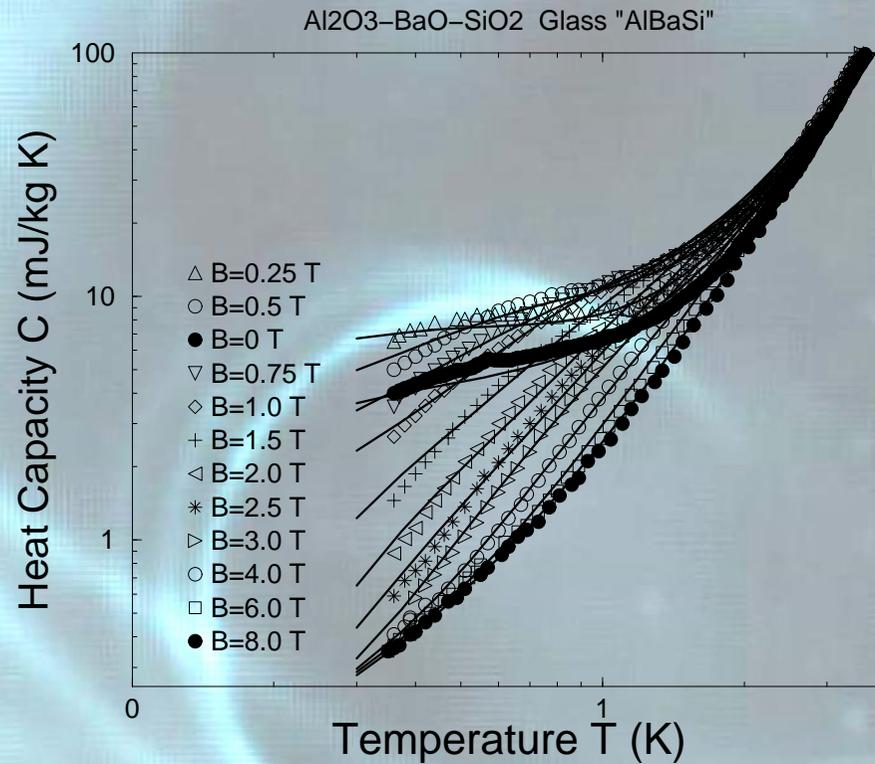
$$\Delta_0 \approx \hbar\Omega e^{-V_B/\hbar\Omega}$$



Situation for Multi-Component Silicate Glasses!

- In a magnetic field, the silicate glass with composition $\text{Al}_2\text{O}_3\text{-BaO-SiO}_2$ shows a surprising dependence on the field B for $T < 1\text{K}$
- Long time ago, this was already observed for the heat capacity (and improperly attributed to paramagnetic impurities, Stephens 1972)
- The heat capacity and the permittivity for mixed $(\text{SiO}_2)_{1-x}(\text{K}_2\text{O})_x$ and $(\text{SiO}_2)_{1-x}(\text{Na}_2\text{O})_x$ glasses are depending on x (at zero field already, MacDonald 1985), never explained

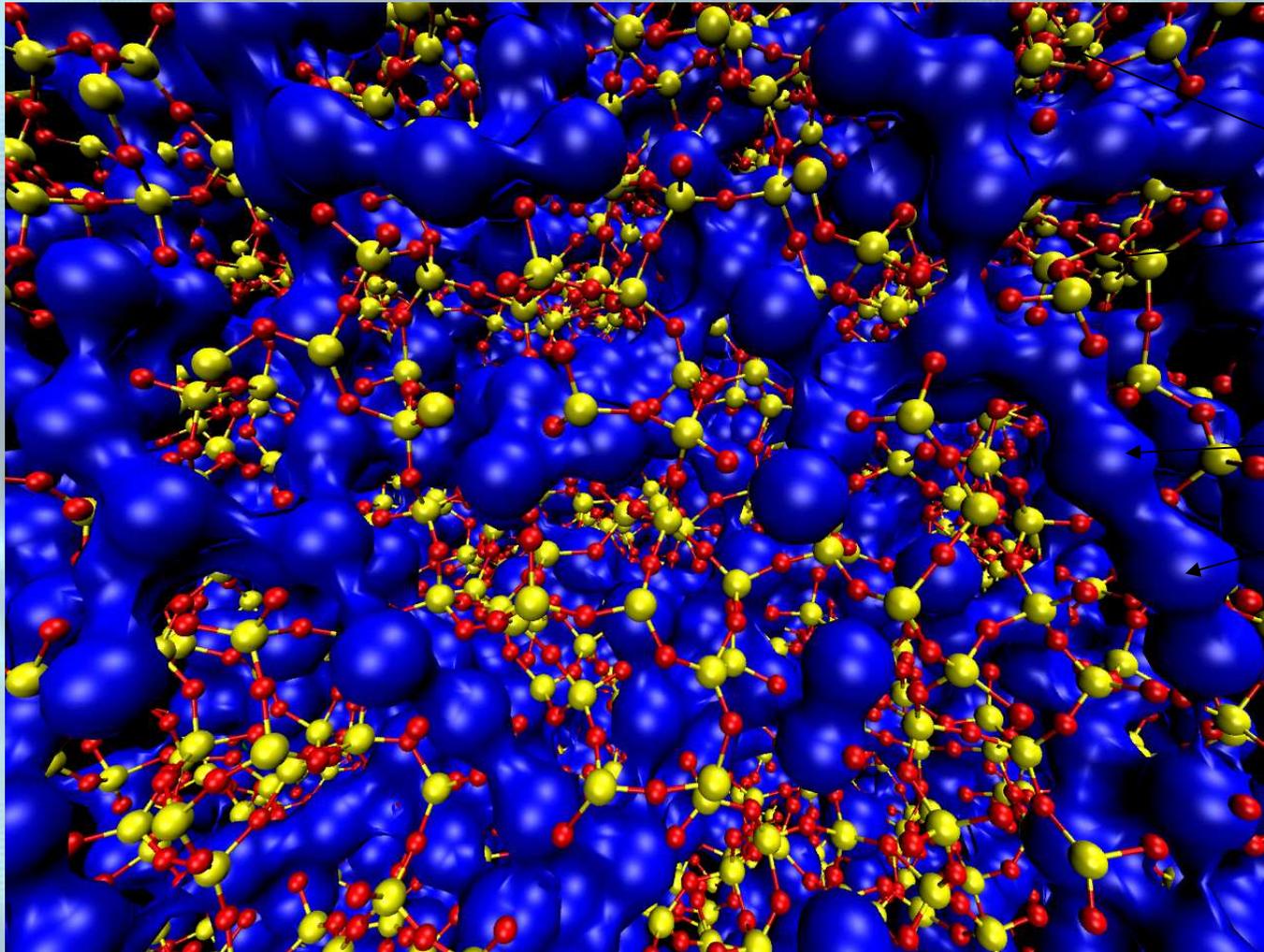
Magnetic field effect



Charged Particle in Multi-Welled Potential

- More than two minima expected (and suggested by MC-simulations of glasses)
- Simulation and experiments show: **multi-component glasses consisting of at least TWO types of “networks”** (Network-Forming (NF) and Network-Modifying (NM))
- NM-pockets can nucleate micro-crystals (observed in some glass films of similar composition)
- Take case of 3-dimensional 3LS with new distribution of parameters for NM tunneling species

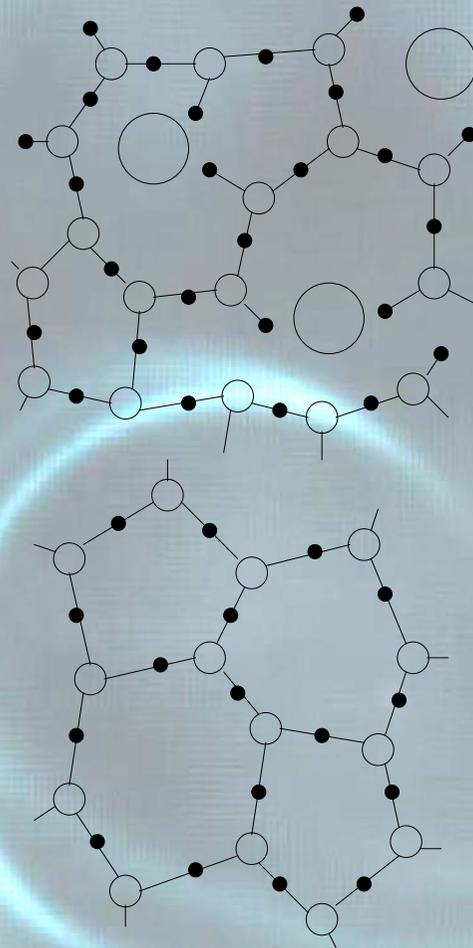
Snapshot of a MD-simulation
(of the glass $(\text{Na}_2\text{O}) \cdot 3(\text{SiO}_2)$ (W.Kob))



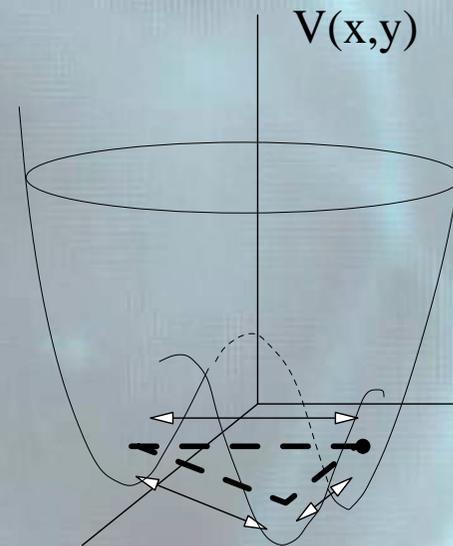
NF
(SiO_4)

NM
(Na^+)

Model Potential for NM Tunneling:



(b)



(c)

(a)



$n_{\text{wells}} > 2$, e.g. **3** for ease of computation

Ordinary glasses (2LS): (tunneling of NF species)

$$H_0 = \frac{1}{2} \begin{pmatrix} -\Delta & \Delta_0 \\ \Delta_0 & +\Delta \end{pmatrix}, \quad \Delta_0 \propto \hbar\Omega e^{-V_0/\hbar\Omega}$$

$$P(\Delta, \Delta_0) = \frac{\bar{P}}{\Delta_0}, \quad \text{uniform - distribution}$$

in Δ and $\ln(\Delta_0)$

pseudo-spin $\frac{1}{2}$ mathematics

Multi-Component Glasses (tunneling of NM species)

$$H_0 = \begin{pmatrix} E_1 & D_0 & D_0 \\ D_0 & E_2 & D_0 \\ D_0 & D_0 & E_3 \end{pmatrix},$$

$$E_1 + E_2 + E_3 = 0,$$

$$D = \sqrt{E_1^2 + E_2^2 + E_3^2}$$

$$P(E_1, E_2, E_3; D_0) = \frac{P^*}{(E_1^2 + E_2^2 + E_3^2)D_0}$$

Dielectric constant (2LS+ATS)

$$\varepsilon = - \left. \frac{\partial^2 f(F)}{\partial F^2} \right|_{F=0}, \quad f(F) = -k_B T \overline{\ln Z}, \quad F - \text{electric field}$$

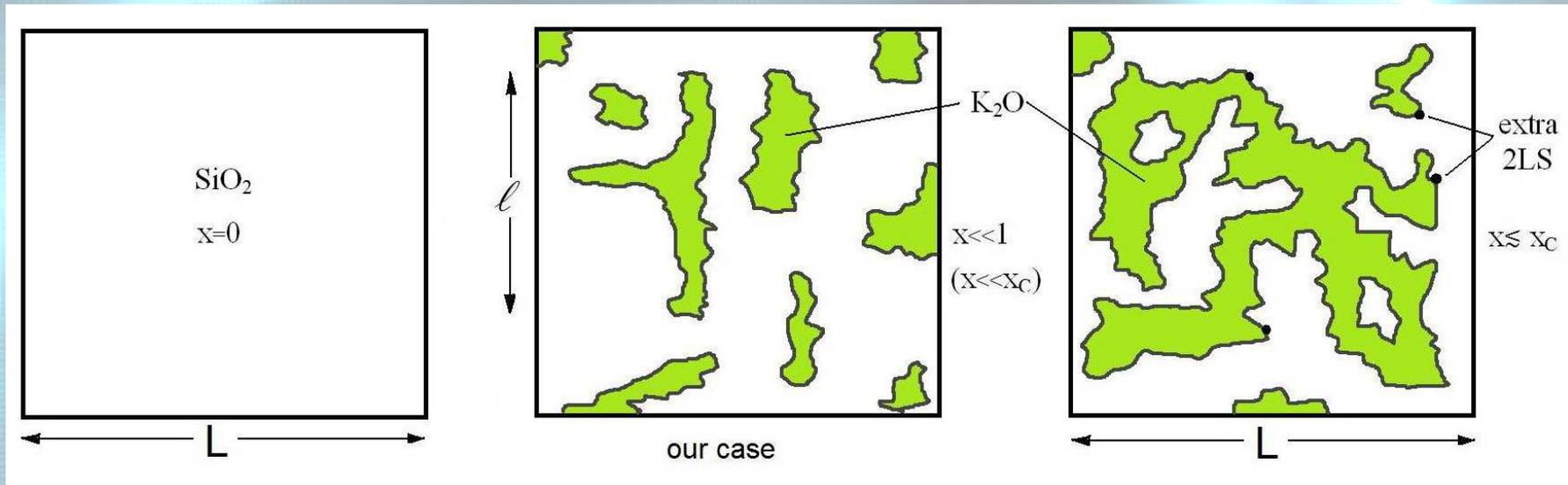
$$\frac{\Delta \varepsilon'(T)}{\varepsilon'} = \left. \frac{\Delta \varepsilon'}{\varepsilon'} \right|_{RES} + \left. \frac{\Delta \varepsilon'}{\varepsilon'} \right|_{REL}$$

$$\left. \frac{\Delta \varepsilon'}{\varepsilon'} \right|_{REL} \propto 1/(1 + \omega^2 \tau^2)$$

$$\tau_{2LS}^{-1} = E \Delta_0^2 / \gamma \tanh\left(\frac{E}{2k_B T}\right), \quad \tau_{ATS_{max}}^{-1} = D^5 / \Gamma \tanh\left(\frac{D}{2k_B T}\right)$$

γ and Γ are some appropriate elastic constants

Percolation in multi-component glass



Near $x=0$:

$$\frac{N_{2LS}^{(s)}}{V} \propto \frac{x P_{\infty}(x) l^{D_s^{(0)}}}{l^{D^{(0)}}} = x P_{\infty}(x) x^{(D_s^{(0)} - D^{(0)}) \nu^{(0)}} \propto x^f$$

$$f = 1 - (D^{(0)} - D_s^{(0)}) \nu^{(0)} < 1$$

B=0 Direct Evidence for 3LS!

Fitting the Data with our Model

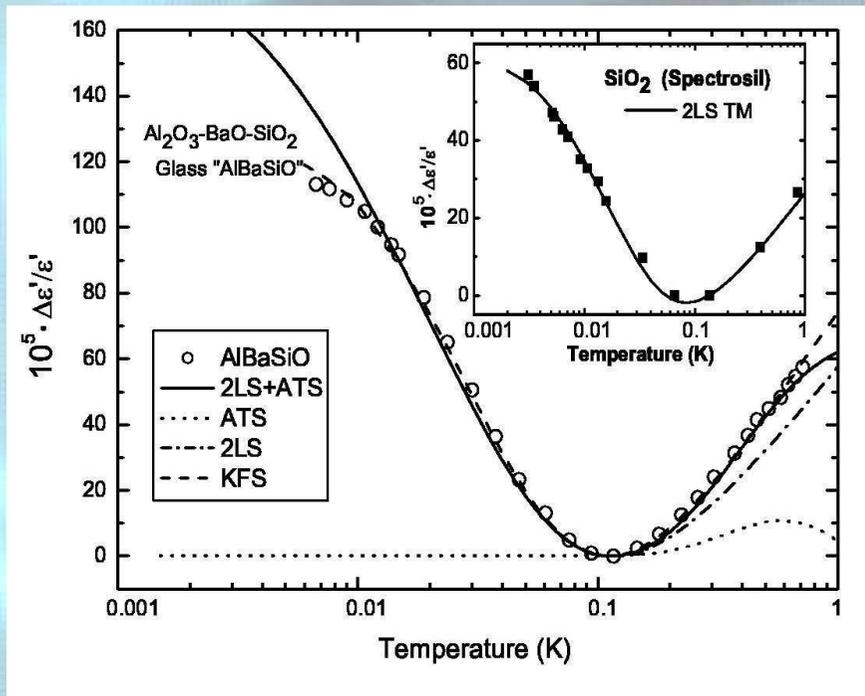


Figure 1 – Dielectric signature of pure α - SiO_2 (inset) and AlBa-SiO (main) glasses.

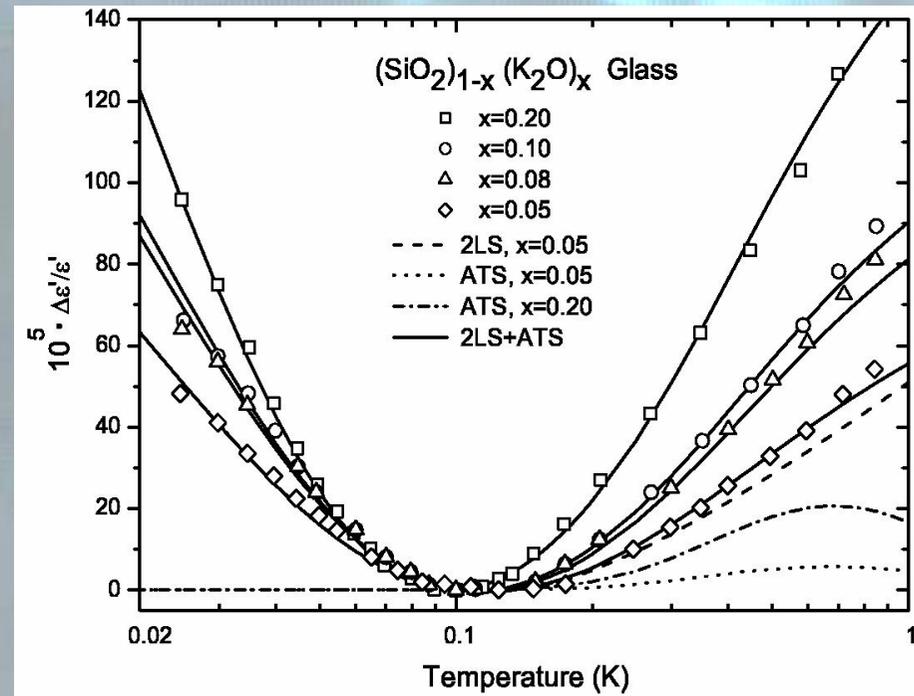


Figure 2 – Dielectric signature of mixed $(\text{SiO}_2)_{1-x}(\text{K}_2\text{O})_x$ glasses as function of T and x .

Data from: G. Frossati (1987), P. Strehlow (1998), W.M. MacDonald (1985)

Potassium-silicate Glass $(\text{SiO}_2)_{1-x}(\text{K}_2\text{O})_x$

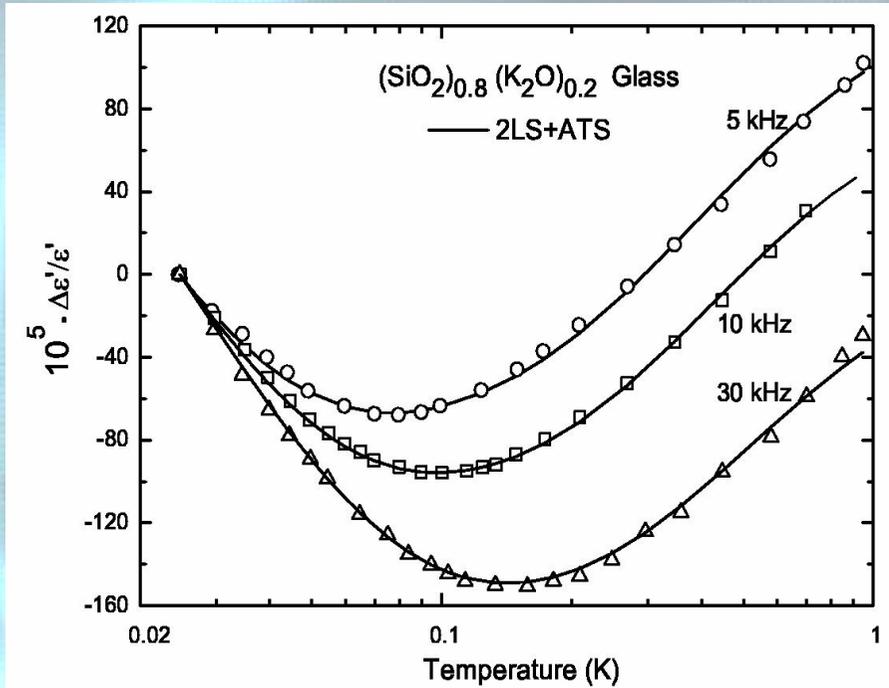


Figure 3 – Dielectric signature of mixed $(\text{SiO}_2)_{1-x}(\text{K}_2\text{O})_x$ glasses as function of T and ω , for $x=0.2$.

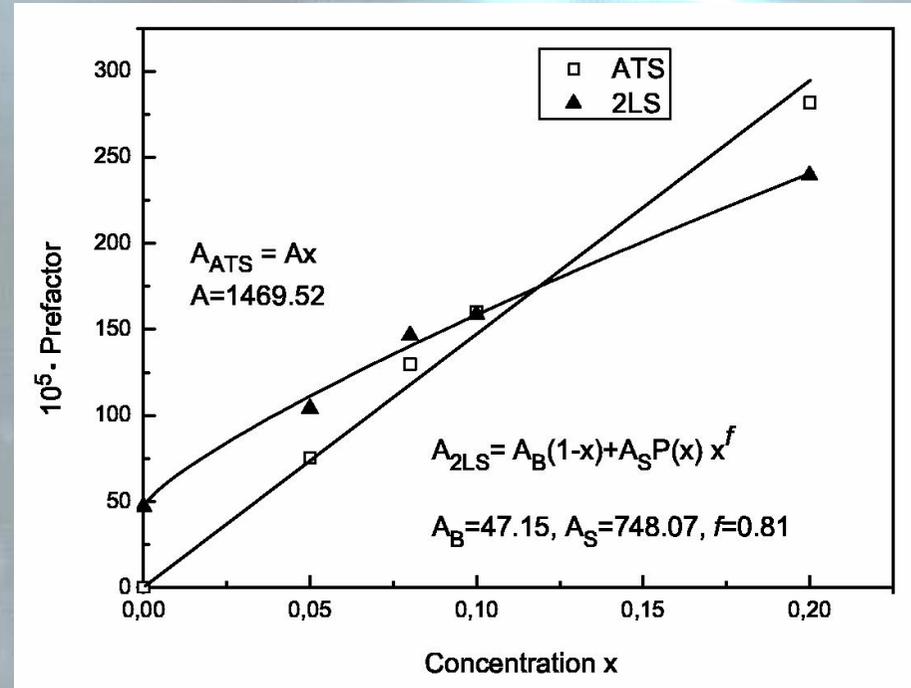


Figure 4 – Dielectric constant prefactors of mixed $(\text{SiO}_2)_{1-x}(\text{K}_2\text{O})_x$ glasses as function of x .

Data from: W.M. MacDonald (1985)

Heat Capacity

$$C_p(T) = B_{ph}T^3 + B_{2LS}T + C_{ATS} + C_p^{Fe^{3+}}$$

$$C_{ATS} = B_{ATS} \left[\ln \left(2 \cosh \left(\frac{D_{\min}}{2k_B T} \right) \right) - \frac{D_{\min}}{2k_B T} \tanh \left(\frac{D_{\min}}{2k_B T} \right) \right]$$

$$C_p^{Fe^{3+}} = B^{Fe^{3+}} \int_0^{\infty} dy \frac{25y^4}{16V_0^3 T^2} e^{-\frac{2y}{V_0}} \cosh^{-2} \left(\frac{5y}{8T} \right)$$

C_{ATS} from our model , $C_p^{Fe^{3+}}$ - from Borisenko (2007)

Results for the Heat Capacity

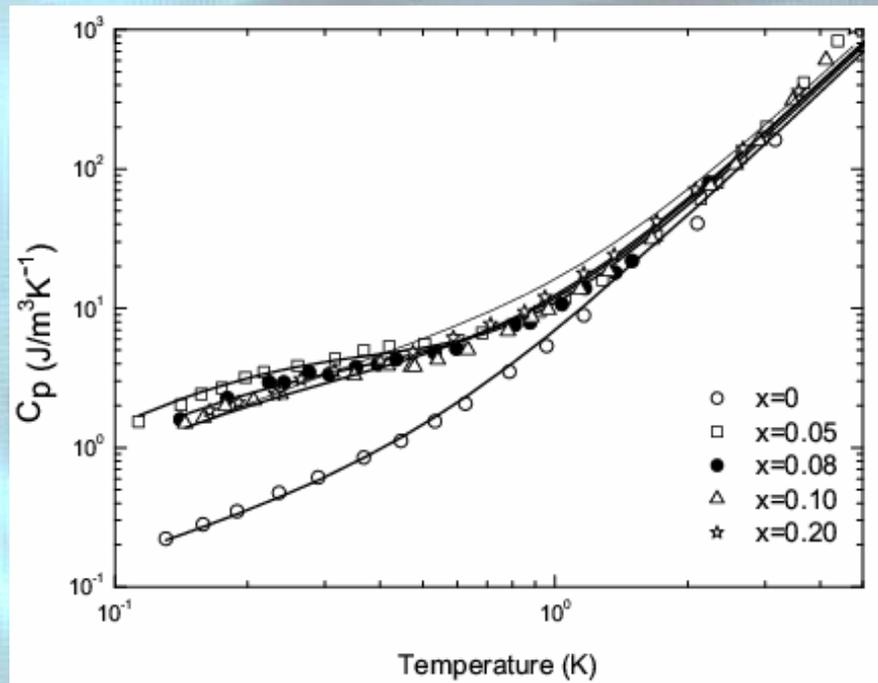


Figure 5 – The heat capacity of mixed $(\text{SiO}_2)_{1-x}(\text{K}_2\text{O})_x$ glasses as function of T and x

Data from: W.M. MacDonald (1985)

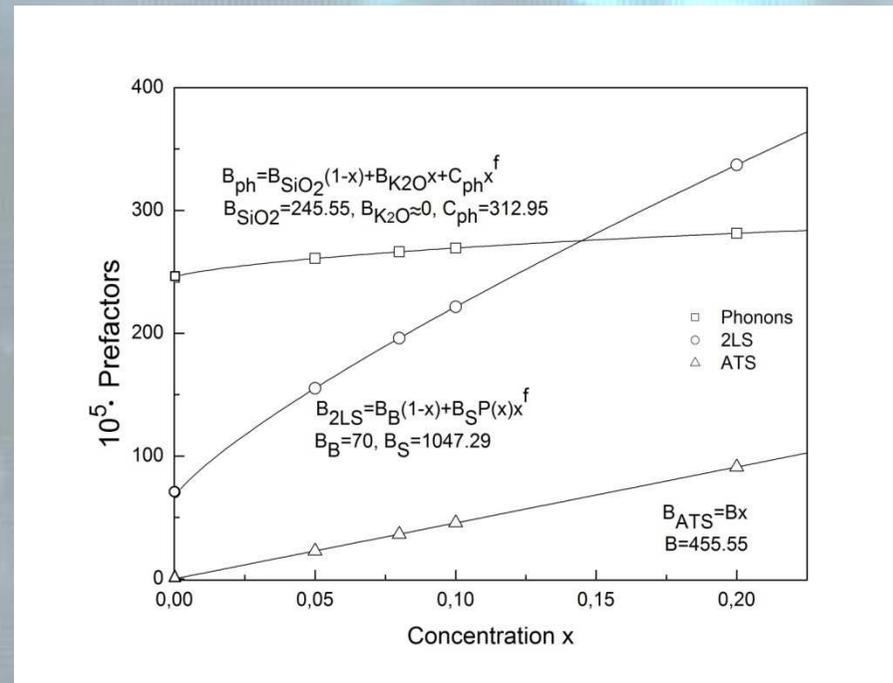


Figure 6 – The prefactor parameters for mixed $(\text{SiO}_2)_{1-x}(\text{K}_2\text{O})_x$ glasses as function of x

CONCLUSIONS

- We have proved the essential role of anomalous multilevel tunneling systems in mixed glasses already at $B=0$
- For $B>0$ these ATS have accounted for the magnetic effect in the heat capacity and dielectric constant of the multi-component glasses
- We plan to extend the theory to the high field regime ($B>1$ T), where more physics is to be explained
- We plan to study the dipole-echo magnetic field dependence in these insulating glasses as well

The background features a dark grey, textured surface with several glowing, ethereal light trails and circles in shades of cyan and white. These light patterns are scattered across the frame, creating a sense of movement and energy. The central text is prominently displayed in a bold, black, sans-serif font.

THANK YOU!