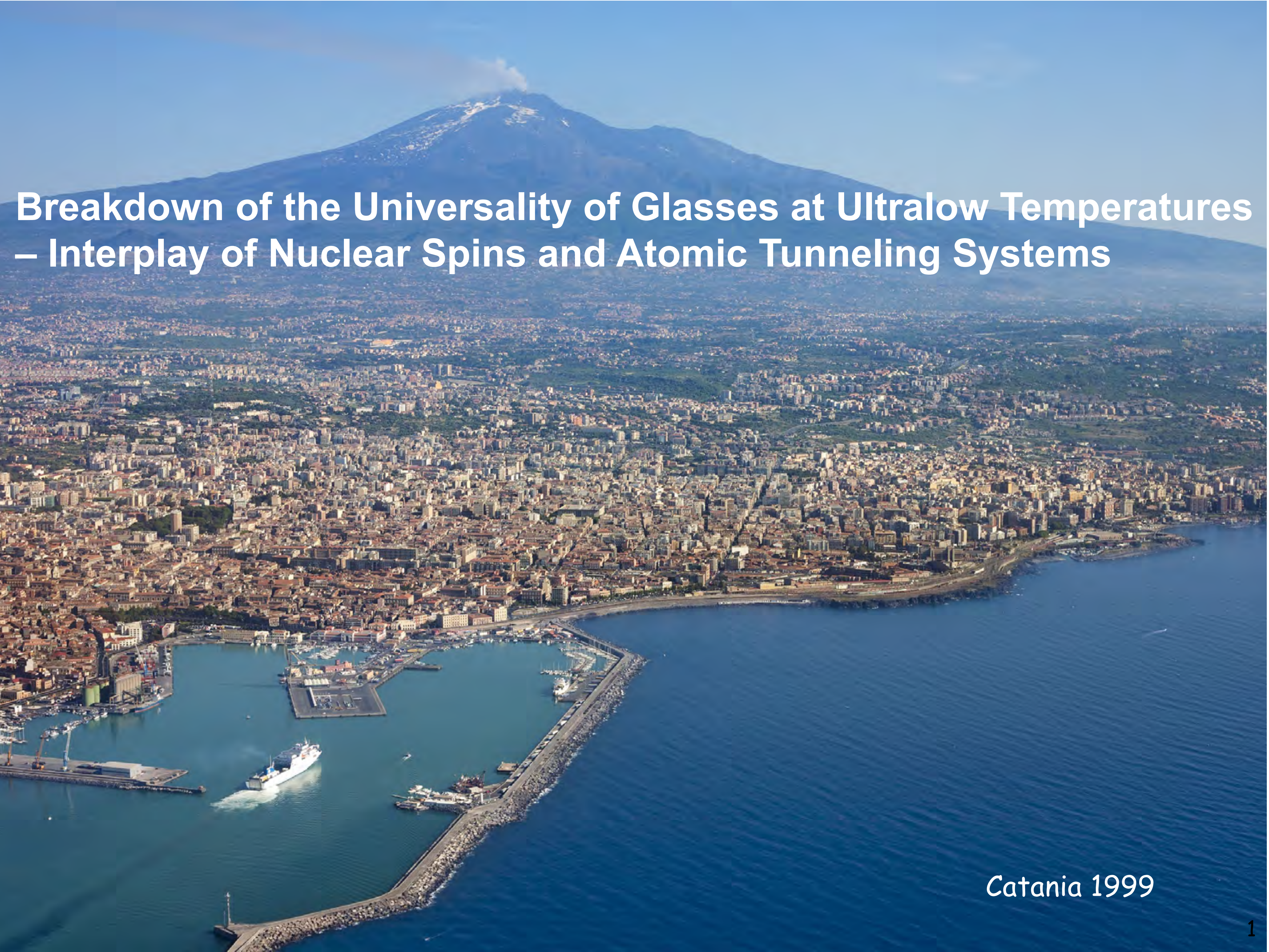
Breakdown of the Universality of Glasses at Ultralow Temperatures – Interplay of Nuclear Spins and Atomic Tunneling Systems



Catania 1999

Comment Paper by Clare and Tony 1988

Low Temperature Properties of Amorphous Materials: Through a Glass Darkly

A wide variety of amorphous materials exhibit similar behavior in their thermal properties. Examples of such universal features include a linear specific heat and a T^2 thermal conductivity below about 1 K. While there is at present no generally accepted model of their behavior at higher temperatures, for the past fifteen years the low-temperature behavior has been attributed to the existence in these materials of tunneling two-level systems. The two-level system model is generally believed to receive strong support from a series of remarkable effects (phonon echoes, etc.) observed in the acoustic behavior of these materials.

In this Comment we point out (a) that while the observed effects are (mostly) consistent with the two-level system model, they do not uniquely establish it, (b) that the model cannot explain the dramatic *quantitative* universality in the low-temperature behavior of amorphous materials, and (c) that the neglect to lowest order within the model of the long-range interactions induced by the strain field, while self-consistent, is not imposed by the data. We sketch an alternative scenario for the properties of amorphous materials in which the role played by these interactions is essential. This scenario, if it can be quantitatively implemented, holds out promise of being able to explain in a unified way not only the quantitative universality below 1 K but also the behavior at higher temperatures.

Comments Cond. Mat. Phys.
1988, Vol. 14, No. 4, pp. 231–251

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Comments Cond. Mat. Phys.
1988, Vol. 14, No. 4, pp. 231–251

… a totally unbelievable degree of chance coincidence

Physica B 169 (1991) 322–327
North-Holland

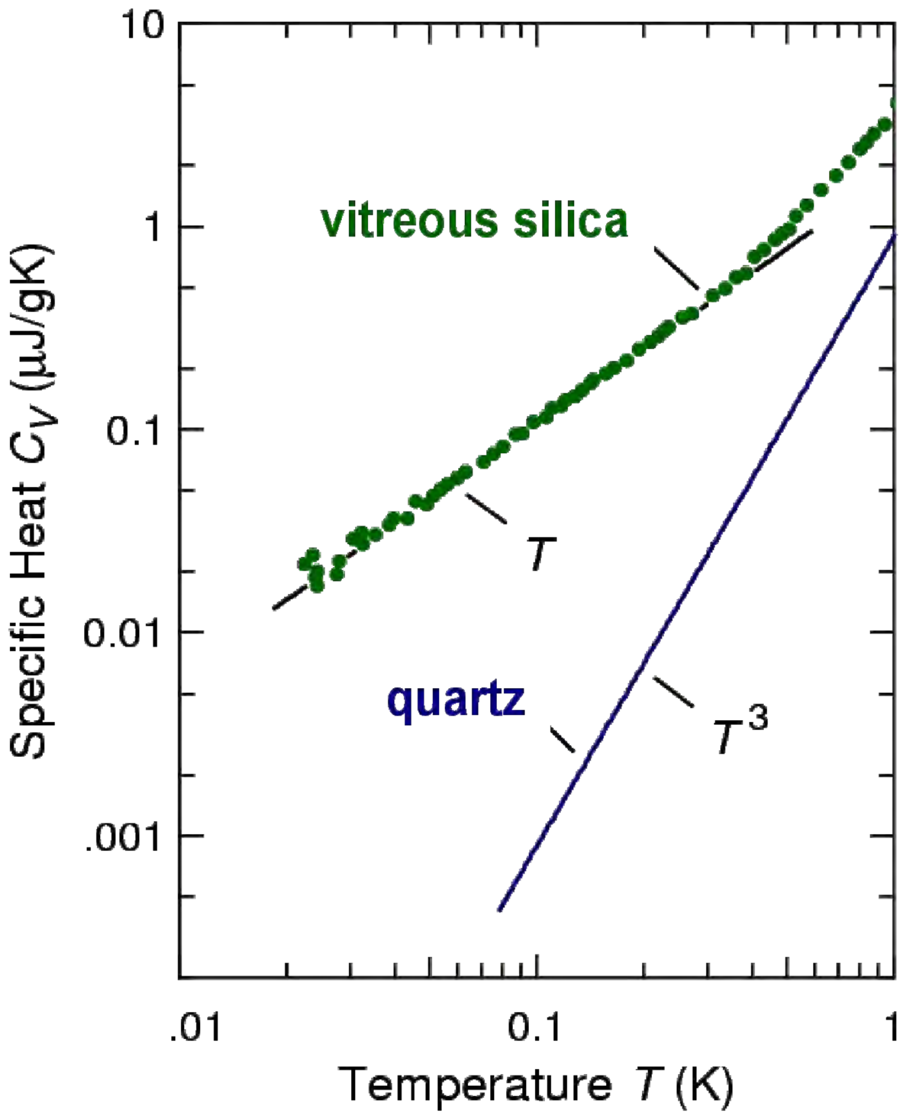
Amorphous materials at low temperatures: why are they so similar?

Anthony J. Leggett
Department of Physics, University of Illinois at Urbana-Champaign, 1110 West Green Street, Urbana, IL 61801, USA

The invited talk was given by the author.

In addition to the well-known qualitative similarity of behavior of a wide range of amorphous materials, one dimensionless quantity, the reduced attenuation of transverse ultrasound, shows a *quantitative* universality whose explanation in terms of the standard "tunnelling two-level system" model would seem to require a totally unbelievable degree of chance coincidence. In addition, while the height of the "plateau" in the thermal conductivity is material-specific, the higher-temperature behavior is consistent with universal behavior. I sketch the outlines of a scenario which holds out hope of understanding these observations.

Specific Heat of Amorphous and Crystalline SiO_2

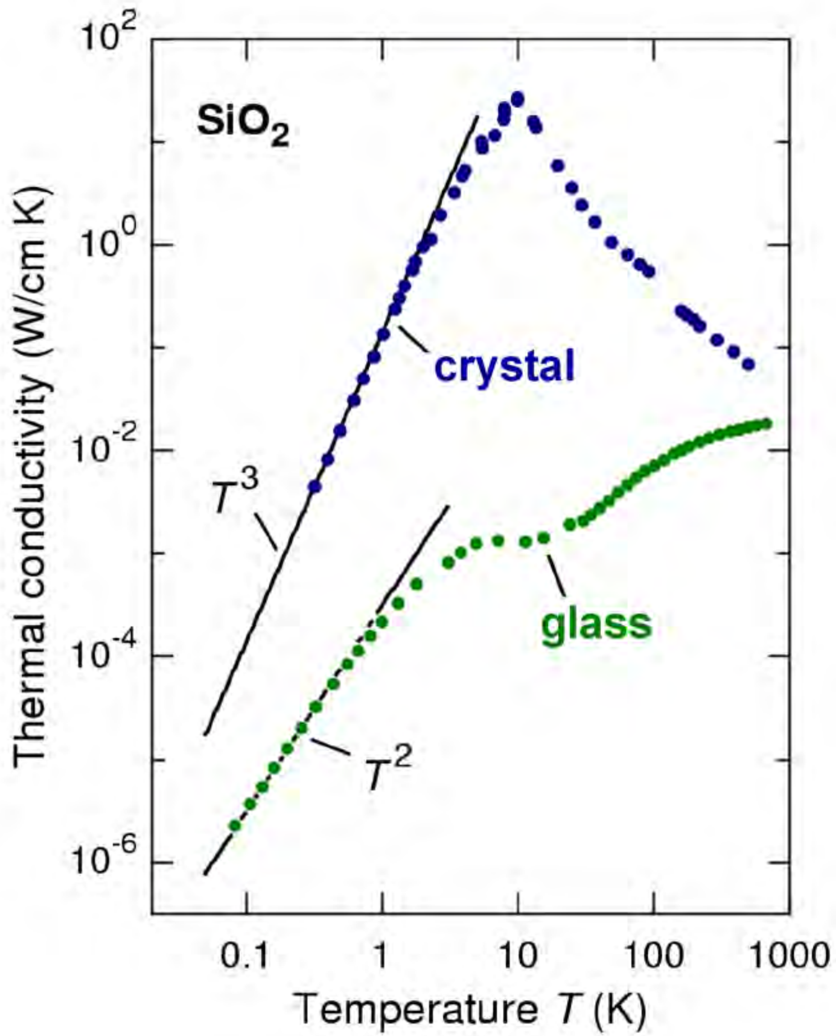


broad distribution of
low-energy excitations

R.C. Zeller, R.O. Pohl,
Phys. Rev. **B 4**, 2029 (1971)

J.C. Lasjaunias et al.,
Sol. State Commun. **17**, 1045 (1975)

Specific Heat of Amorphous and Crystalline SiO_2

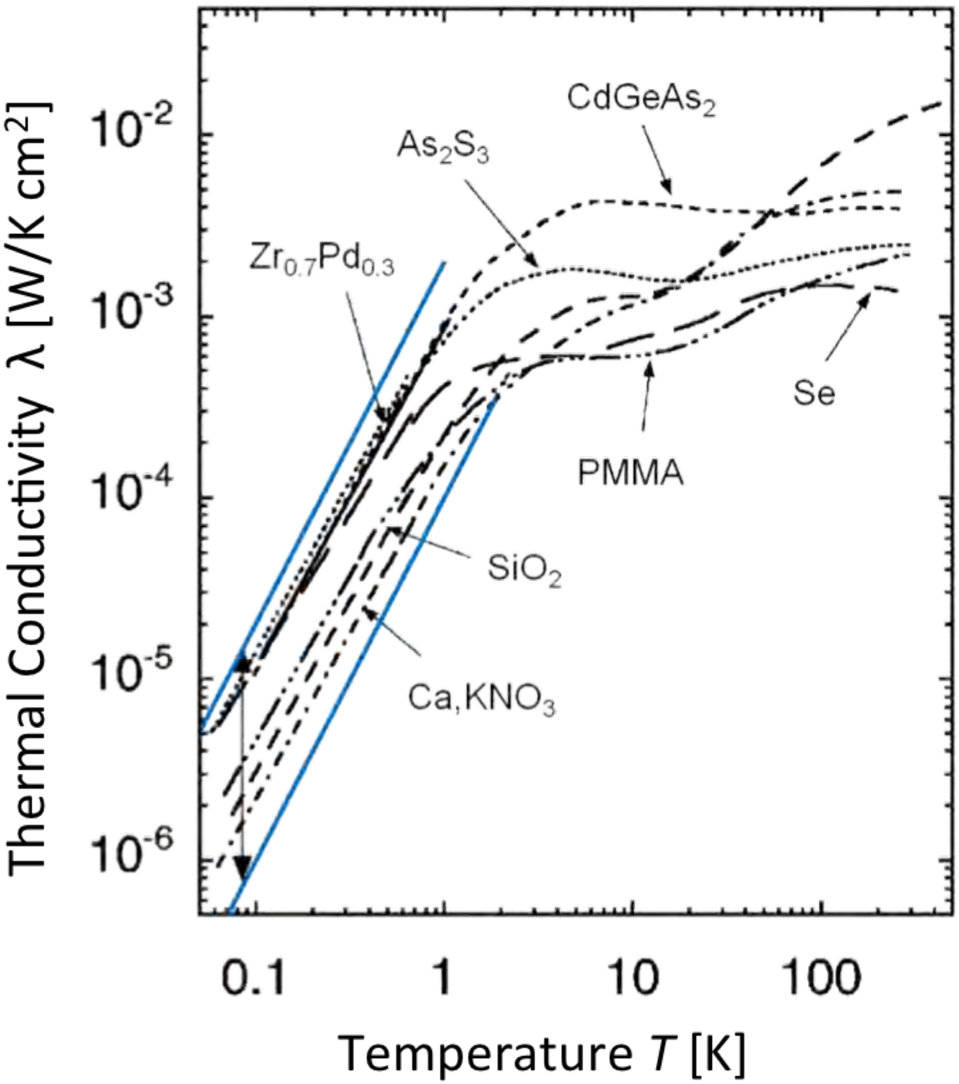


➡ excitations are localized

➡ strong coupling to phonons

R.C. Zeller, R.O. Pohl,
Phys. Rev. **B 4**, 2029 (1971)

Universality of Glasses

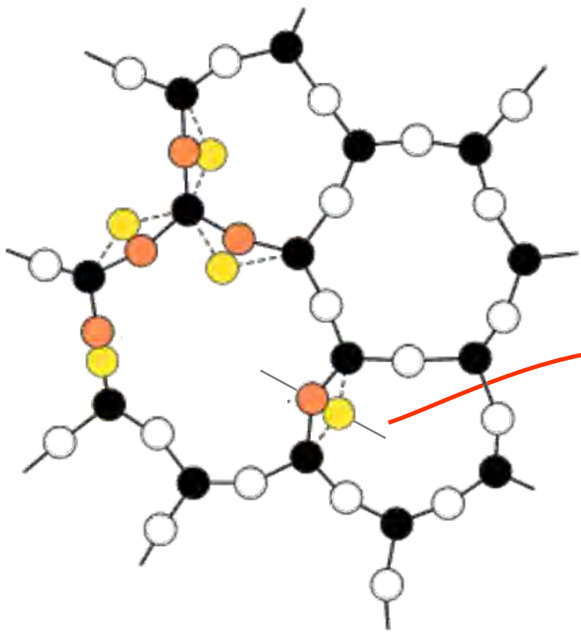


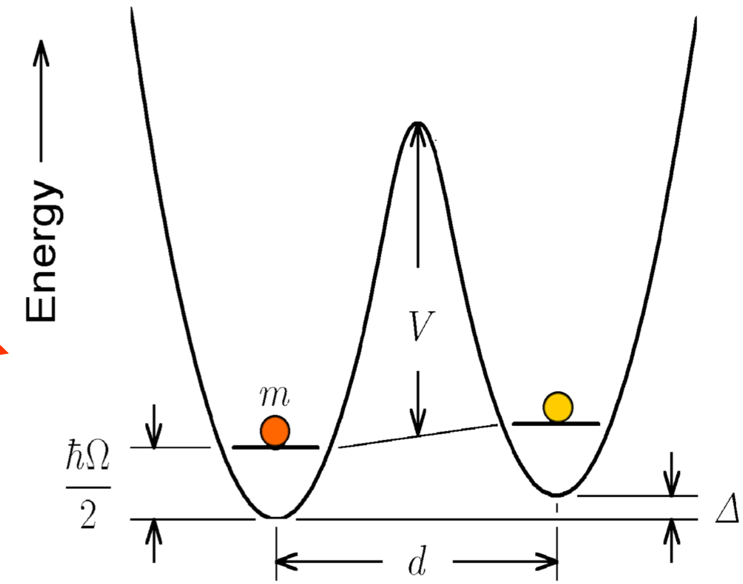
Thermal conductivity of glasses within one order of magnitude

R. B. Stephens, Phys. Rev. B **8**, 2896 (1973)

Atomic Tunneling Systems in Glasses

W.A. Phillips, J. Low. Temp. Phys. **7**, 351 (1972)
P.W. Anderson et al., Philos. Mag. **25**, 1 (1972)





energy splitting

$$E = \sqrt{\Delta_0^2 + \Delta^2}$$

tunnel splitting

$$\Delta_0 = \hbar\Omega\, e^{-\lambda}$$

$$\lambda = \frac{d}{2\hbar}\sqrt{2mV}$$

distribution function

$$P(\lambda, \Delta)\, d\lambda\, d\Delta = \overline{P}\, d\lambda\, d\Delta$$

elastic, dielectric und thermal properities


Dresden 2003


bibliothek

Is there an unambiguous test for TLS in glasses?

"Tunneling Two-Level Systems" Model of the Low-Temperature Properties of Glasses: Are "Smoking-Gun" Tests Possible?

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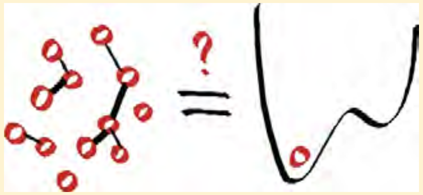
†Department of Physics, University of Illinois at Urbana–Champaign, 1110 West Green Street, Urbana, Illinois, United States

‡Applied Physics, SEAS, Harvard University, 29 Oxford Street, Cambridge, Massachusetts, United States

ABSTRACT: Following a brief review of the "two-level (tunneling) systems" model of the low-temperature properties of amorphous solids ("glasses"), we ask whether it is in fact the unique explanation of these properties as is usually assumed, concluding that this is not necessarily the case. We point out that (a) one specific form of the model is already experimentally refuted and (b) that a definitive test of the model in its most general form, while not yet carried out, would appear to be now experimentally feasible.



J. Phys. Chem. B 2013, 117, 12966–12971

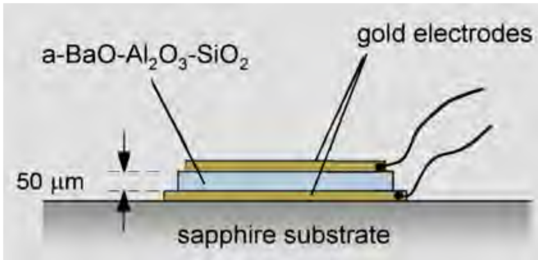
The Cindarella Problem of Glasses

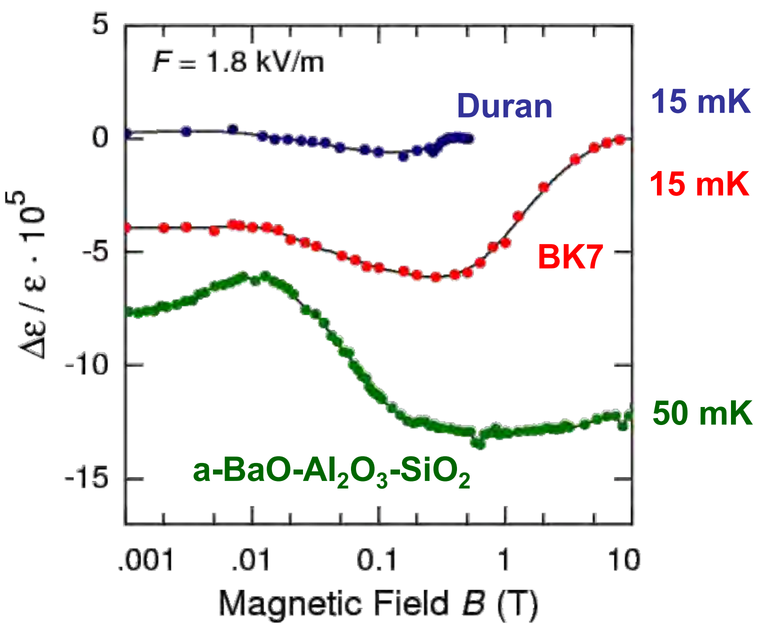
Universality of glasses cannot be explained with the Standard TLS Model (although it is itself no contradiction to it).

At the same time universality is the very reason, why it is so hard to determine the microscopic nature of the excitations in glasses.

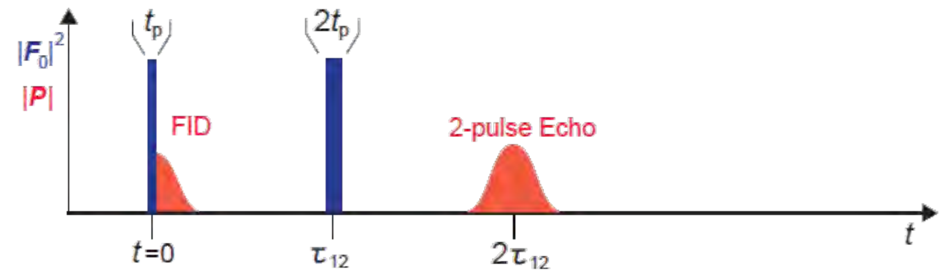
Unexpected Magnetic Field Dependence

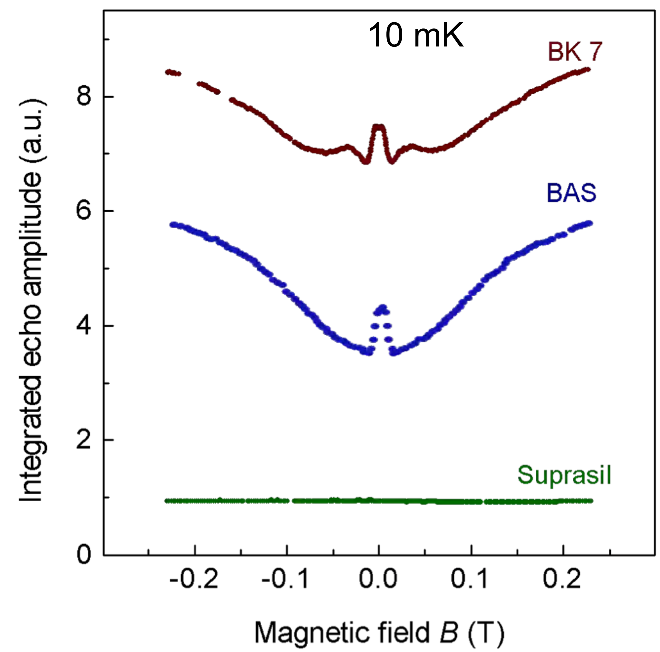
Dielectric Susceptibility





Dipole Echoes



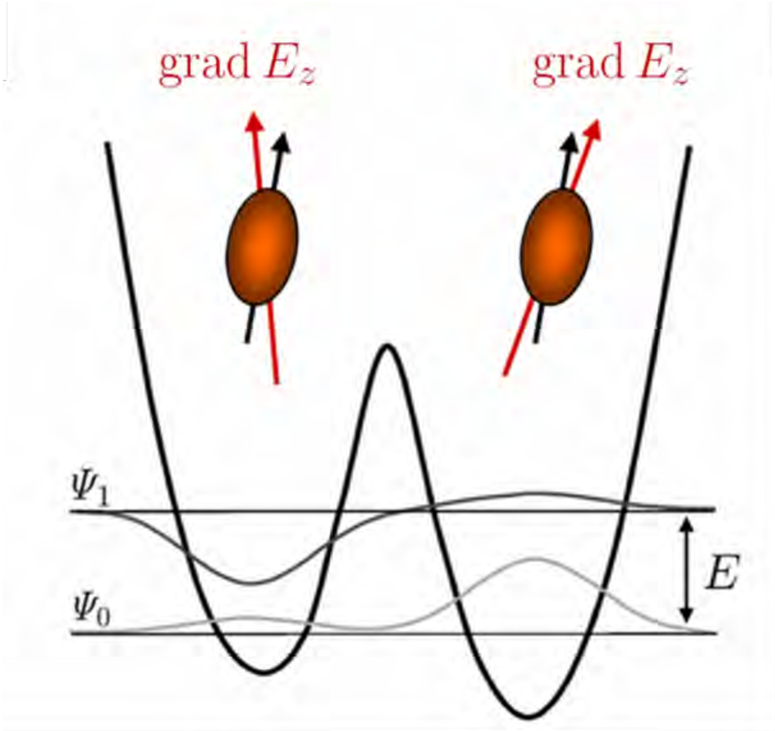


BK7 and Duran: M. Wohlfahrt, P. Strehlow, C. E., S. Hunklinger, Europhys. Lett. **56**, 690 (2001)

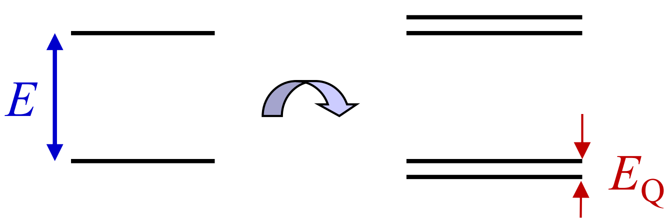
a-BaO- Al$_2$O$_3$- SiO$_2$: R. Haueisen, G. Weiss, Physica B **316-317**, 555 (2002)

S. Ludwig. C. E., S. Hunklinger, P. Strehlow, Phys. Rev. Lett. **88**, 75501 (2002)

Nuclear Quadrupole Moment is Important



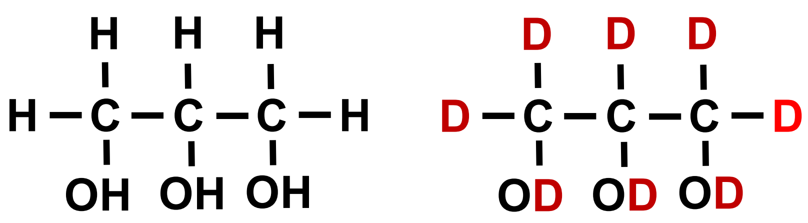
- nuclear quadrupole moment sees electric field gradient in the two wells
  - → hyperfine splitting of tunneling levels
  - → multi-level systems
- magnetic field causes an additional Zeeman splitting of nuclear levels
- no effect for a-SiO_2 because no quadrupole moment

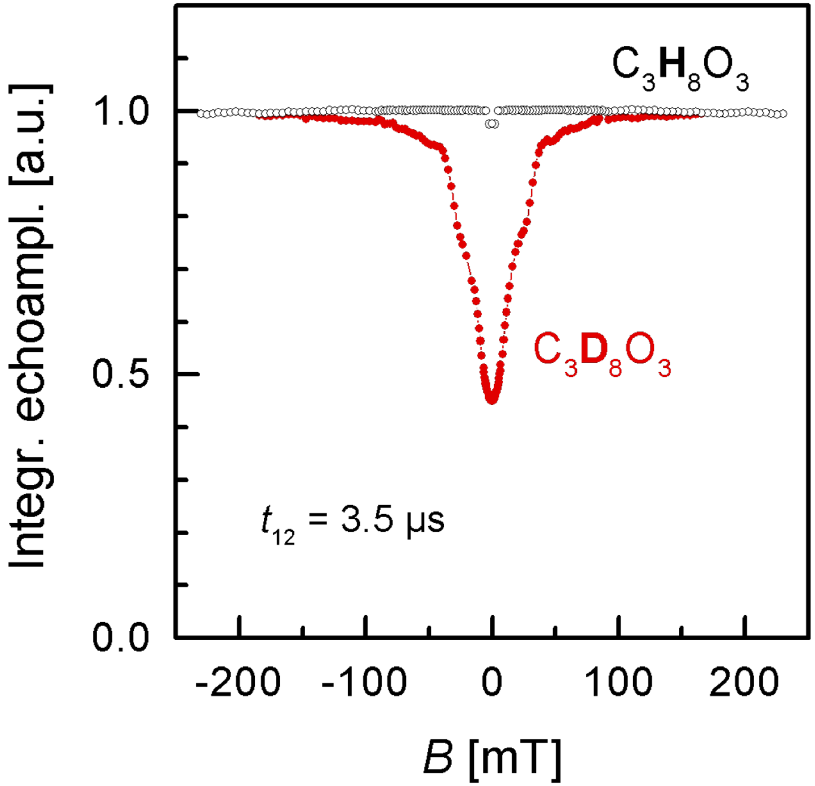


A. Würger, A. Fleischmann, C. E., Phys. Rev. Lett. **89**, 237601 (2002)

Proof: Isotope Effect H $\leftrightarrow$ D

Glycerol





hydrogen

$I = 1/2, \quad \mu = 2.79\,\mu_{\rm N}, \quad Q = 0$

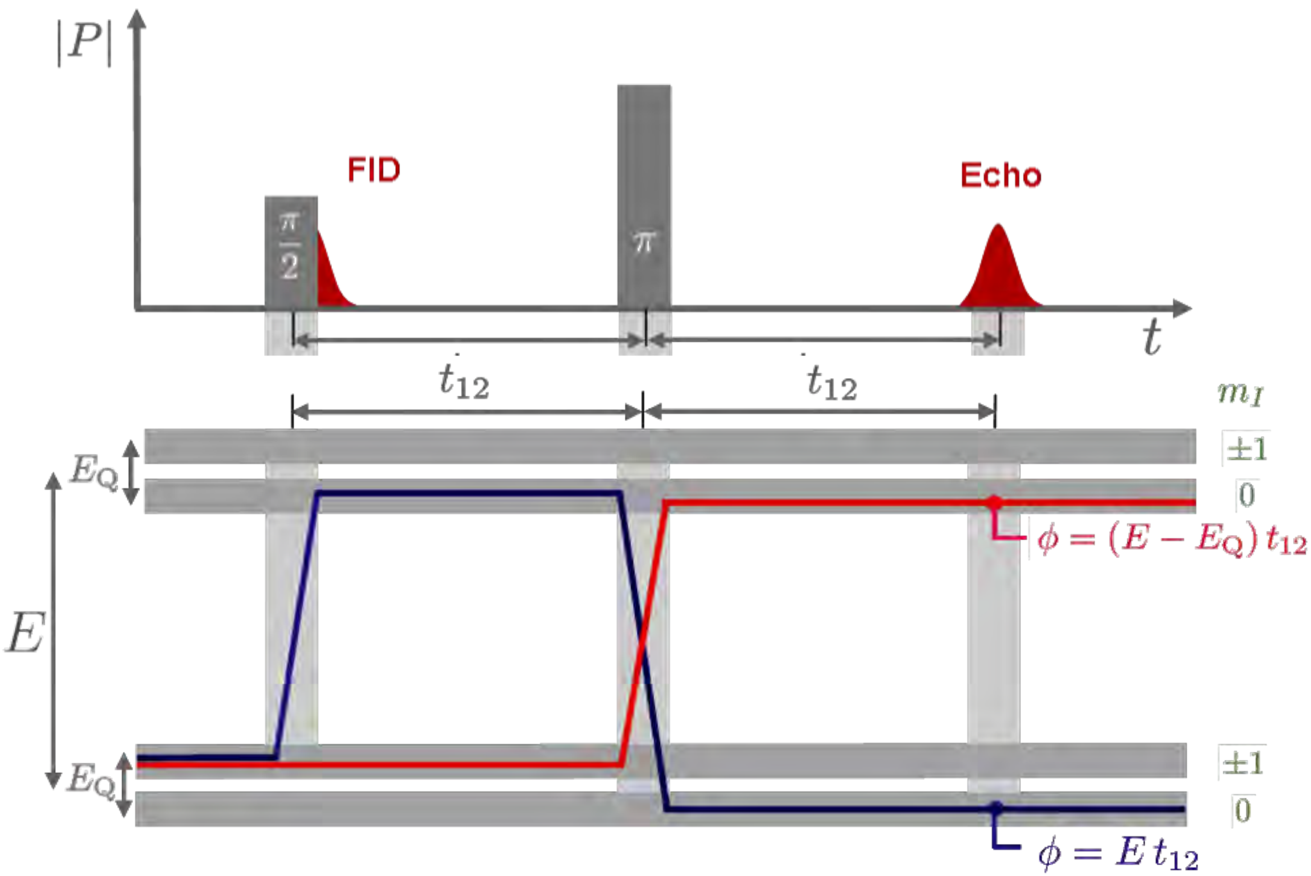
deuterium atom

$I = 1, \quad \mu = 0.86\,\mu_{\rm N}, \quad Q = 0.0029\,b$

P. Nagel, A. Fleischmann, S. Hunklinger, C. E., Phys. Rev. Lett. **92**, 245511-1 (2004)

$\Rightarrow$ proof of the quadrupole model

Zero Magnetic Field



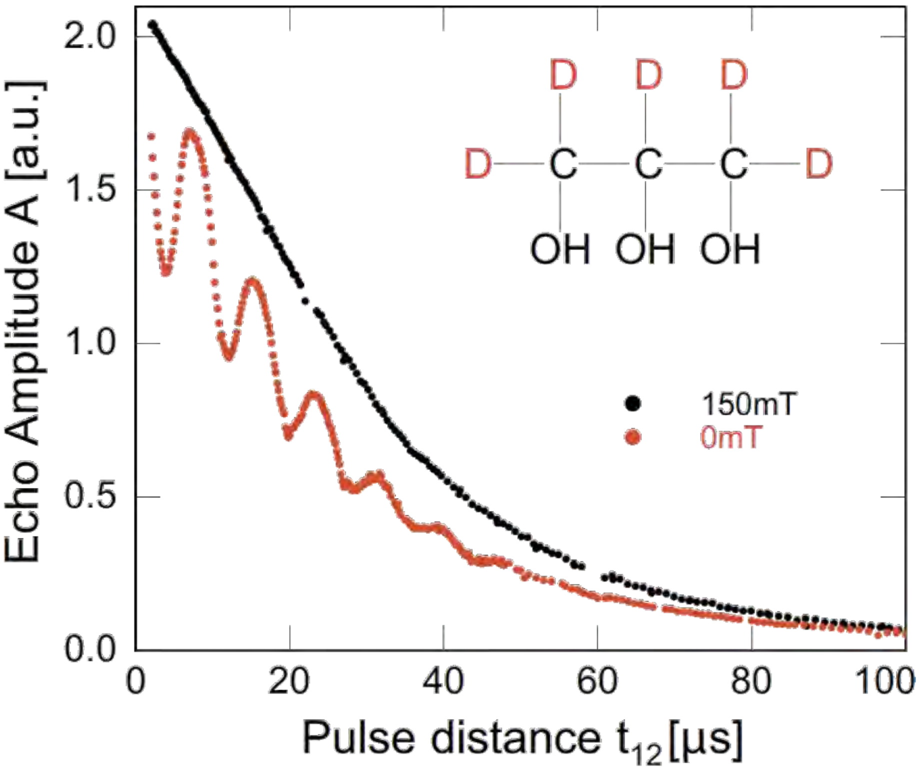
A. Würger, JLTP **137**, 143 (2004)

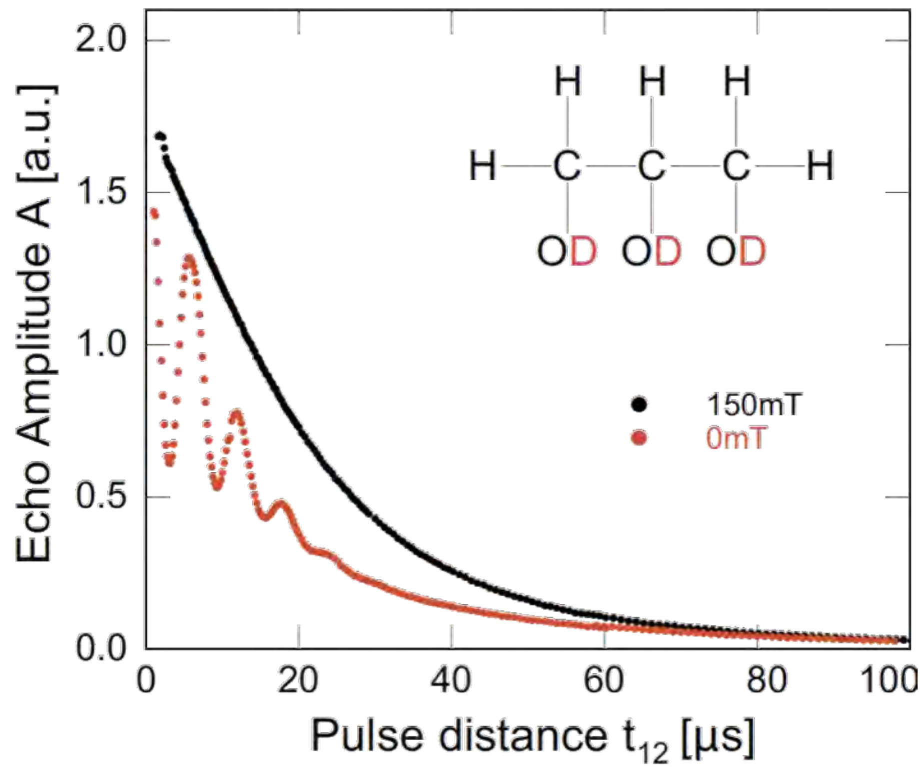
D.A. Parshin, JLTP **137**, 233 (2004)

$$A = A_0 \left[a_1 + a_2 \cos(\omega_Q t_{12}) + a_3 \cos(2\omega_Q t_{12}) \right]$$

$$a_1 + a_2 + a_3 = 1$$

Partially Deuterated Glycerol





$\nu_Q = 128\,\mathrm{kHz}$

$\nu_Q = 125\,\mathrm{kHz}$ (NMR)

$\nu_Q = 160\,\mathrm{kHz}$

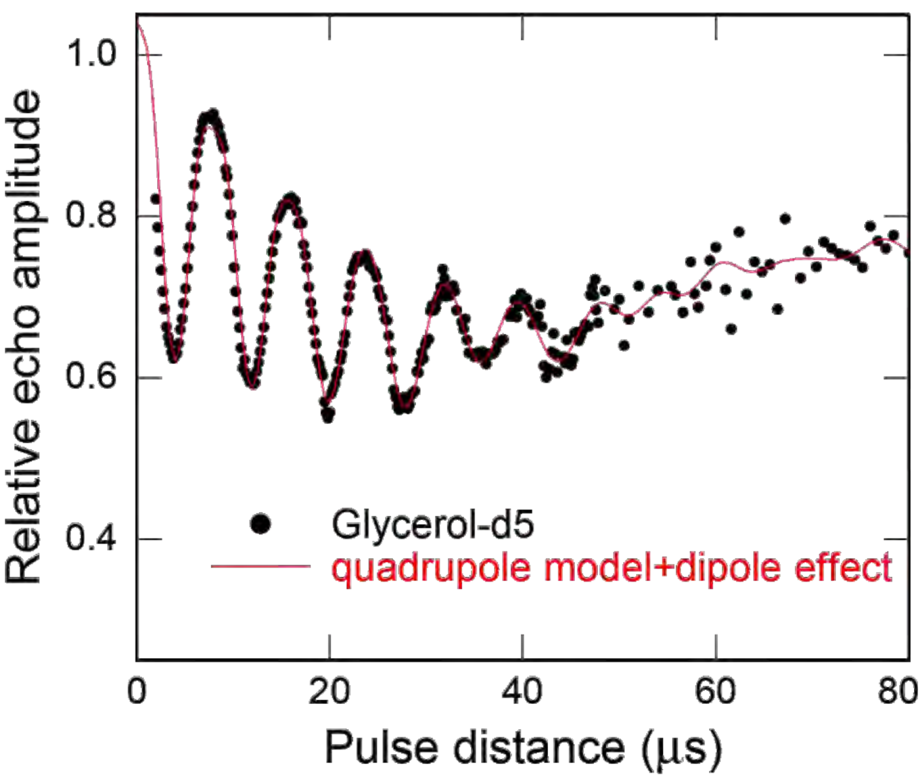
$\nu_Q = 158\,\mathrm{kHz}$ (NMR)

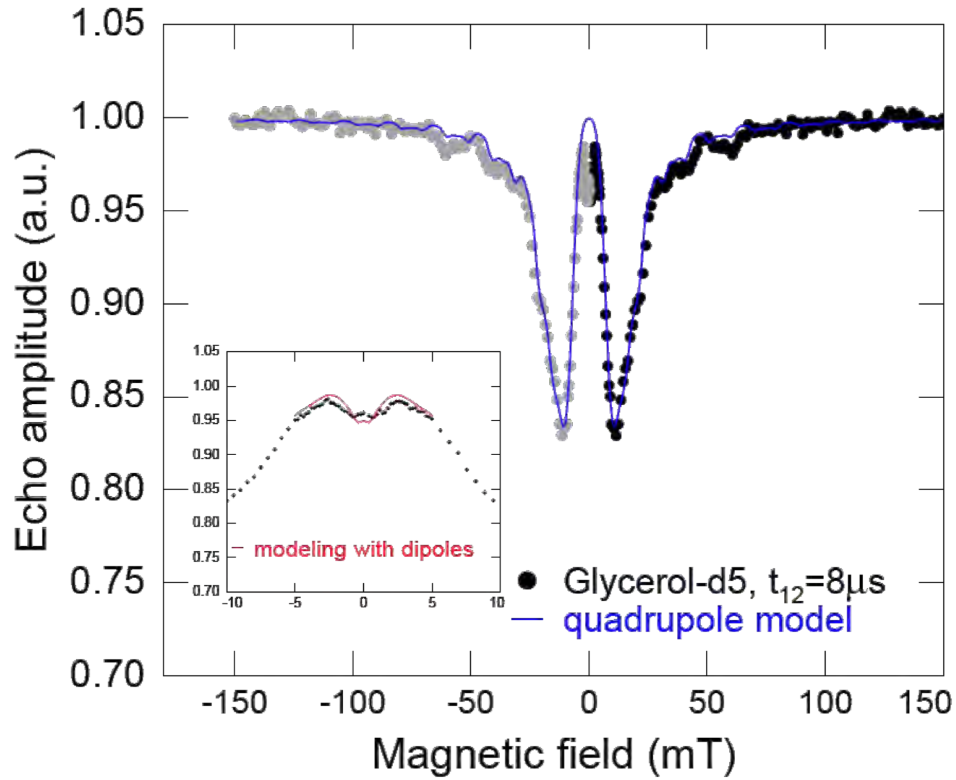
W. Schnauss, F. Fujara, H. Sillescu, J. Chem. Phys. **97**, 1378 (1992).

Beating in case of d3 disappears *faster* → local enviroment ?

A. Bartkowiak, M. Bazrafshan, C. Fischer, A. Fleischmann, C. E., Phys. Rev. Lett. **110** (2013)

Microscopic Modeling





Tunneling angle $2\Theta = 17^\circ$

M. Bazrafshan, PhD Thesis 2008

A. Bartkowiak, M. Bazrafshan, C. Fischer, A. Fleischmann, C. E., Phys. Rev. Lett. **110** (2013)

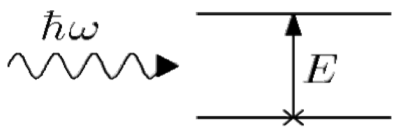
First measurement of microscopic properities of TLS in Glass

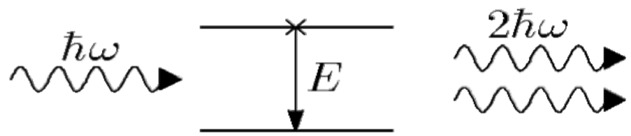
Dielectric Susceptibility

Can nuclear spins explain the magnetic field dependence of the dielectric susceptibility?

Coupling to Electric and Elastic Fields

resonant processes



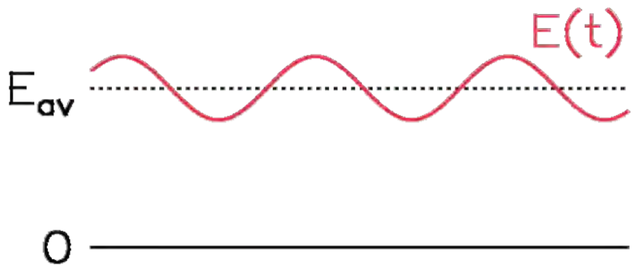


relaxational processes

$\longrightarrow$ modulation of Δ

$$\delta\Delta = 2\gamma e$$

$$\delta\Delta = 2\,\boldsymbol{p}\cdot\boldsymbol{F}$$

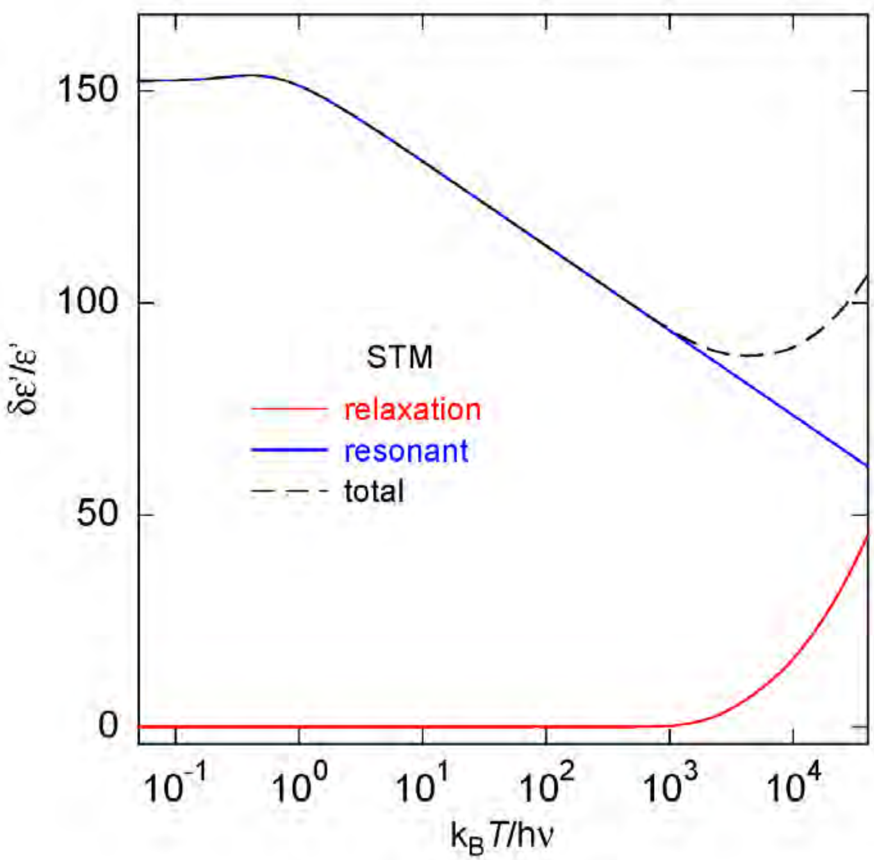


$T < 1$ K one-phonon relaxation

$$\tau_1 = A\left(\frac{E}{\Delta_0}\right)^2 \frac{1}{E^3}\tanh\left(\frac{E}{2k_\mathrm{B}T}\right)$$

Dielectric Susceptibility in the STM

Real part after integration of tunnling parametern distribution



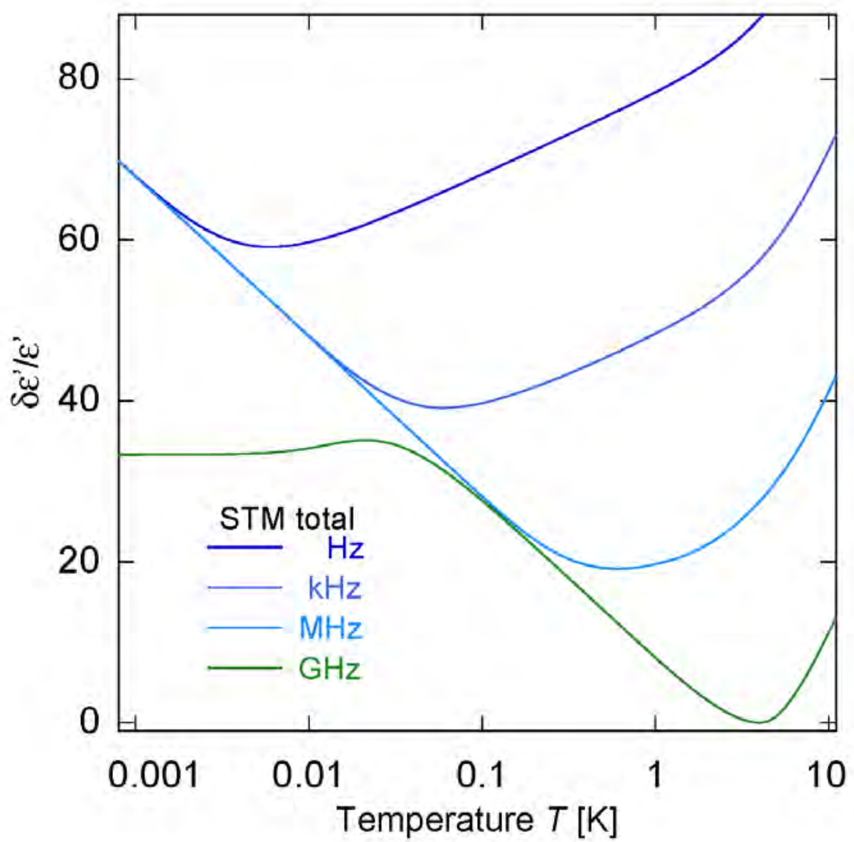
Different Frequencies

Relaxation Rate

$$\tau_{\rm 1ph}^{-1} \propto E^3 \propto T^3 \longrightarrow T_{\rm min} \propto f^{1/3}$$ one phonon

$$\tau_{\rm tot}^{-1} = \tau_{\rm 1ph}^{-1} + \tau_{\rm 2ph}^{-1}$$

$$\tau_{\rm 2ph}^{-1} \propto T^7 \longrightarrow T_{\rm min} \propto f^{1/7}$$ two phonon

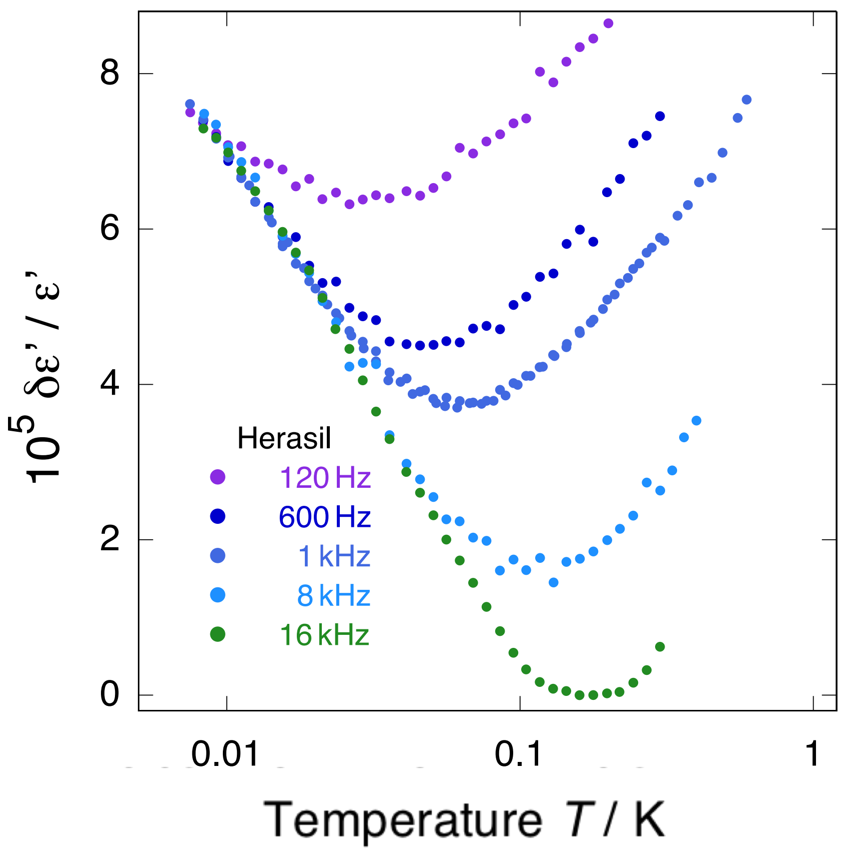


Samples

Name	Type of Glass	Quadrupole Moment Q
CVAc	Polymer glass	None
Herasil	Pure quartz glass	None
BK7	Multi-component glass	No large Q
N-KZFS11	Multi-component glass	^{181}Ta, Q =3.3 barn
HY-1	Multi-component glass	^{165}Ho, Q =3.5 barn

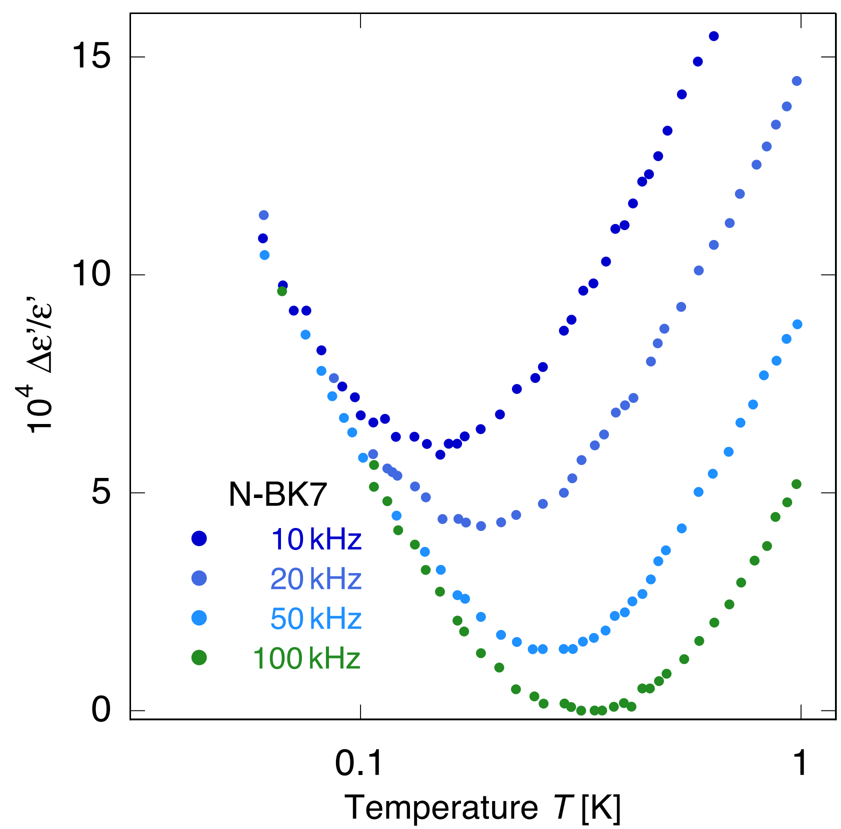
Dielectric Susceptibility of Standard Glasses

Herasil



A. Luck, A. Fleischmann, A. Reiser, C. E., J. Phys. C **568**, 032013 (2014)

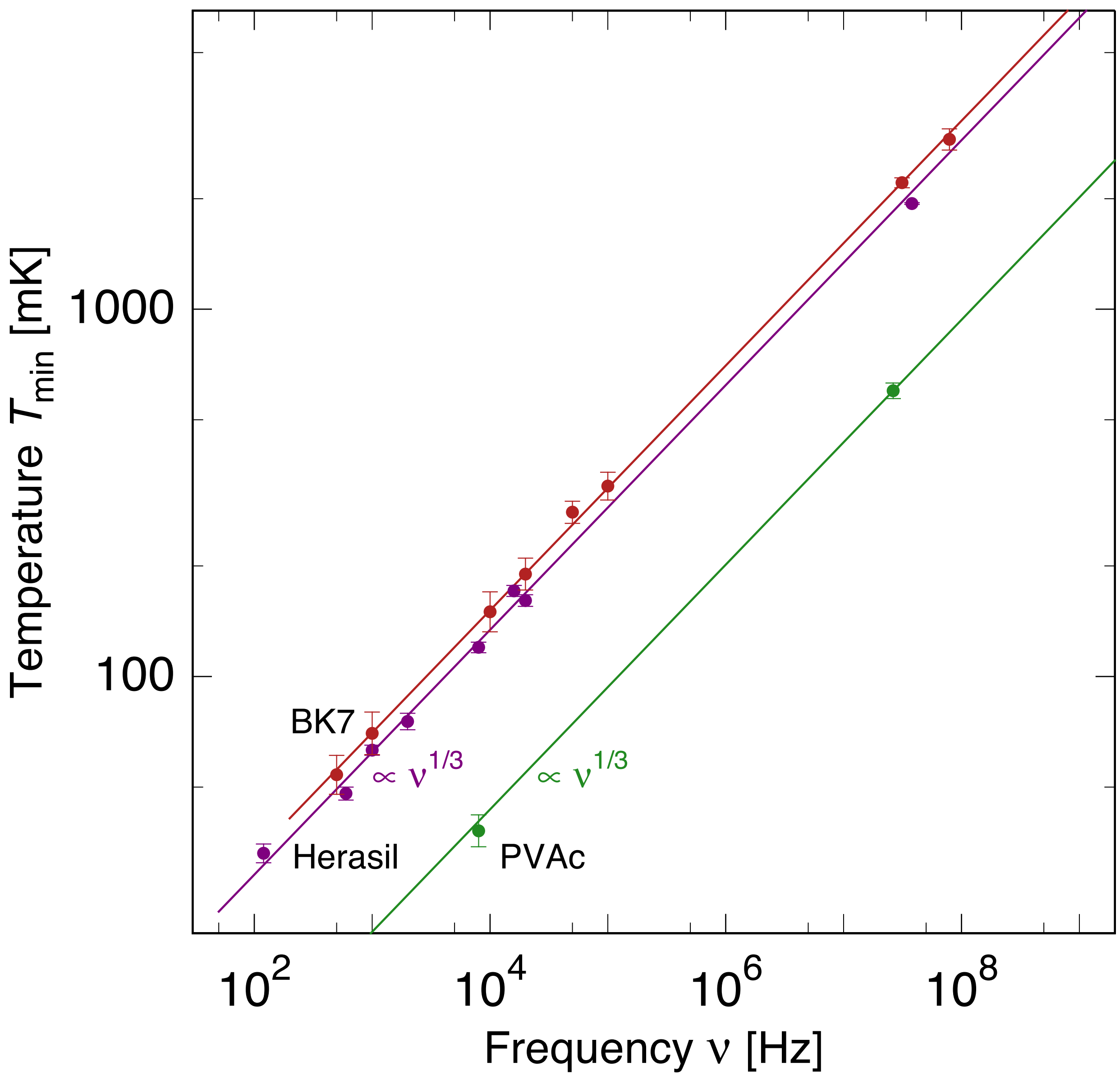
Multi-component glass BK7



C.E., C. Bechinger, M. v. Schickfus, Phonons **89**, 474 (1989)

- Logarithmic temperature dependence
- shift of minimum temperature with frequency

Frequency Dependence of Minimum in DK



one phonon process:

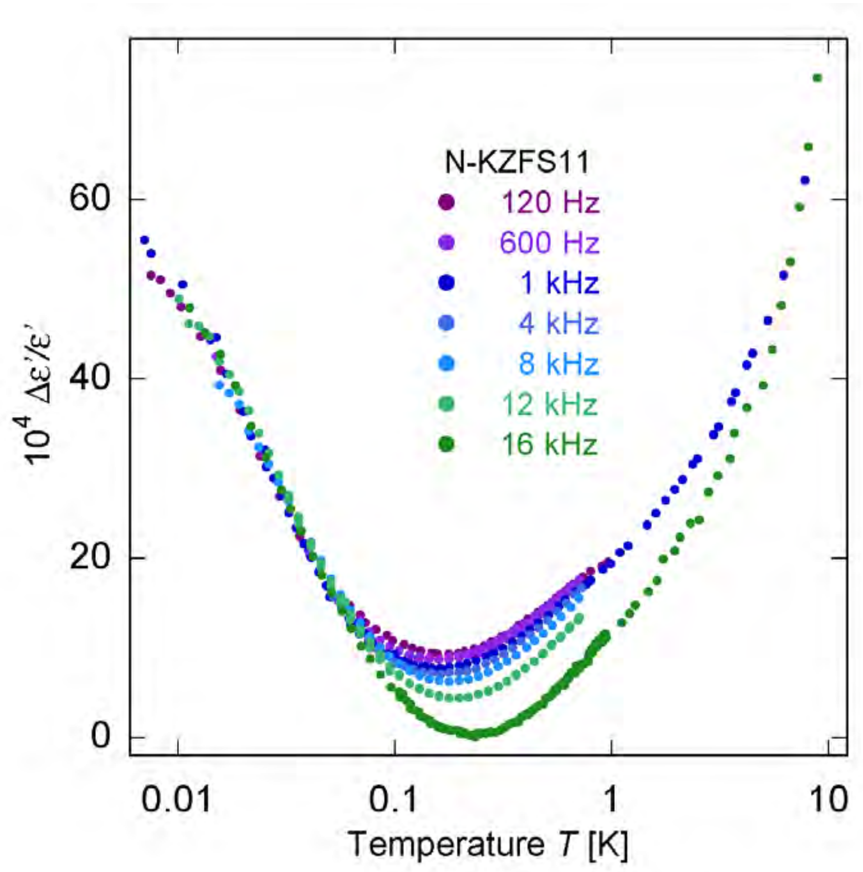
$$T_{\min} \propto \nu^{1/3}$$

two phonon process:

$$T_{\min} \propto \nu^{1/7}$$

Dielectric Susceptibility of Glasses With Nuclear Quadrupols

Multi-component glass containing Tatalum

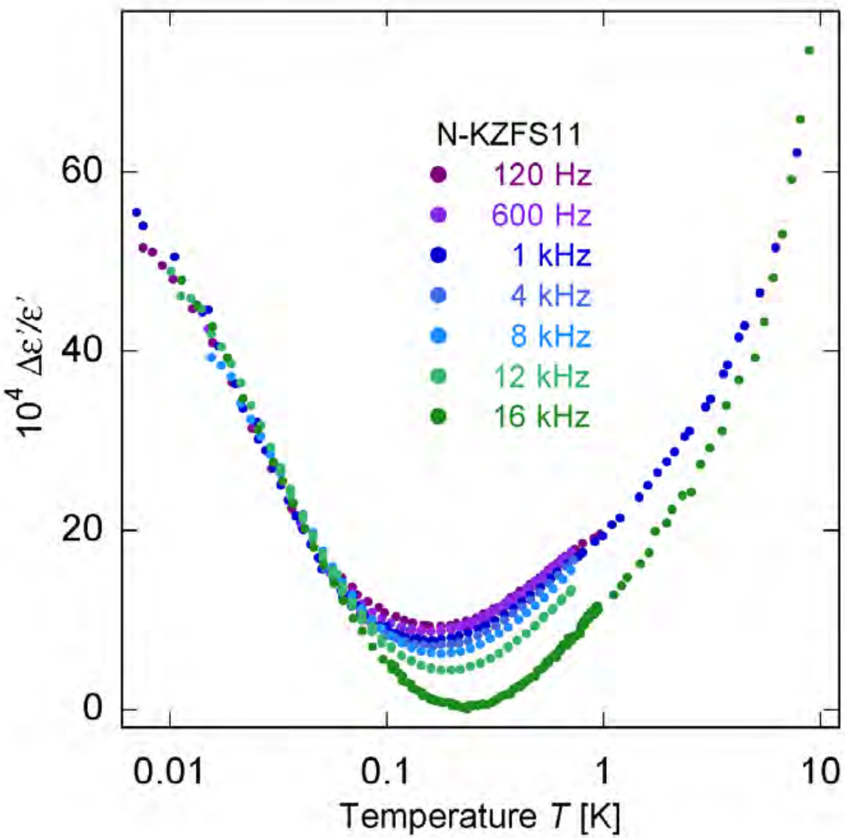


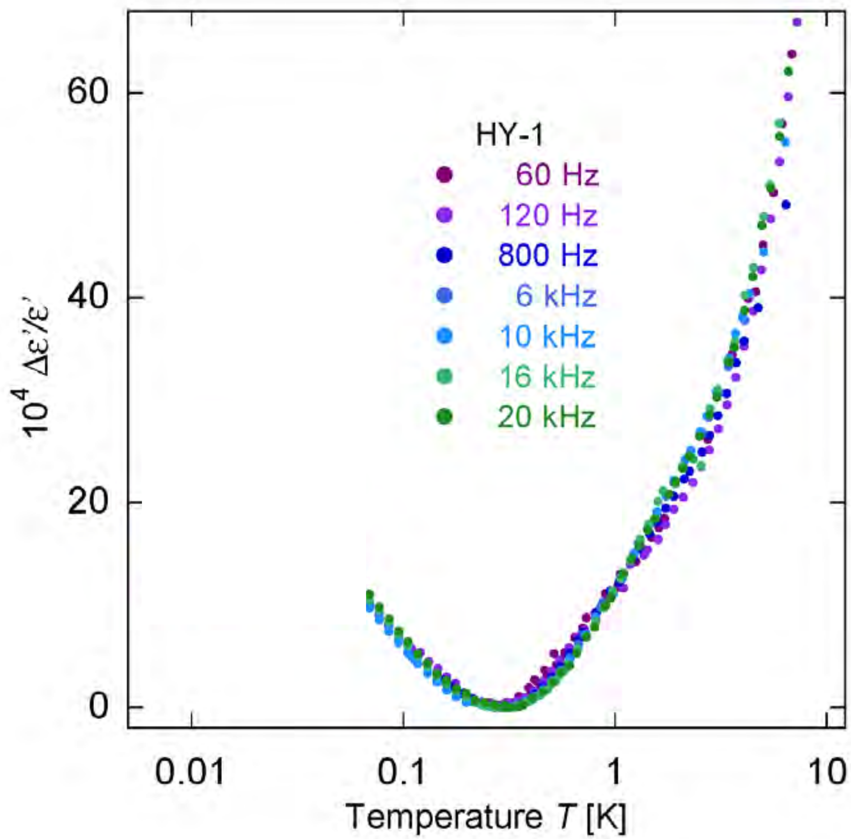
A. Luck, A. Fleischmann, A. Reiser, C. Enss, J. Phys. C **568**, 032013 (2014)

Dielectric Susceptibility of Glasses With Nuclear Quadrupols

Multi-component glass containing Tatalum

Multi-component glass containing Holmium

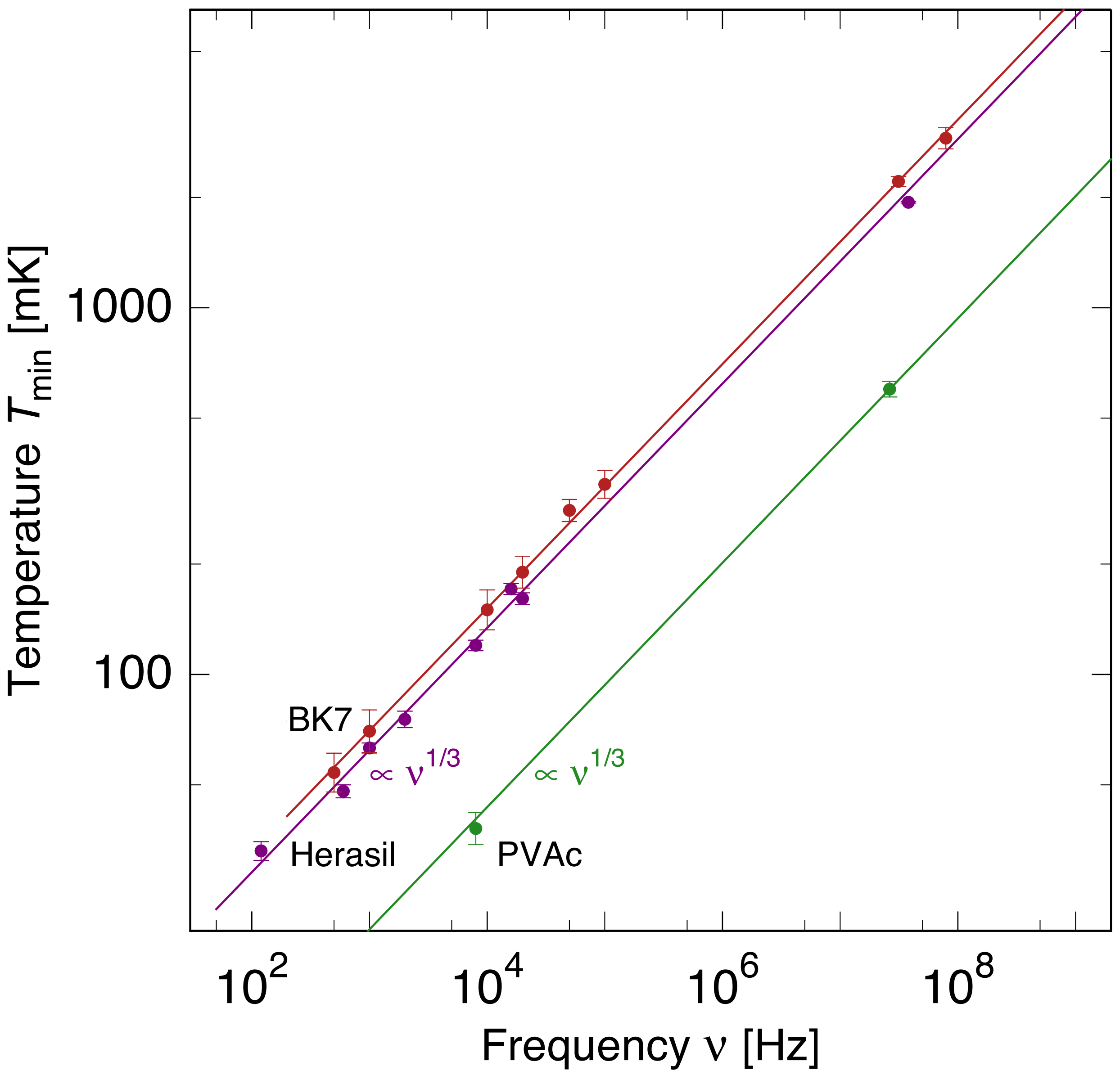




A. Luck, A. Fleischmann, A. Reiser, C. Enss,
J. Phys. C **568**, 032013 (2014)

- HY-1 and N-KZFS11 different from other glasses
- no shift of minimum temperature with frequency at low frequencies

Frequency Dependence of Minimum in DK



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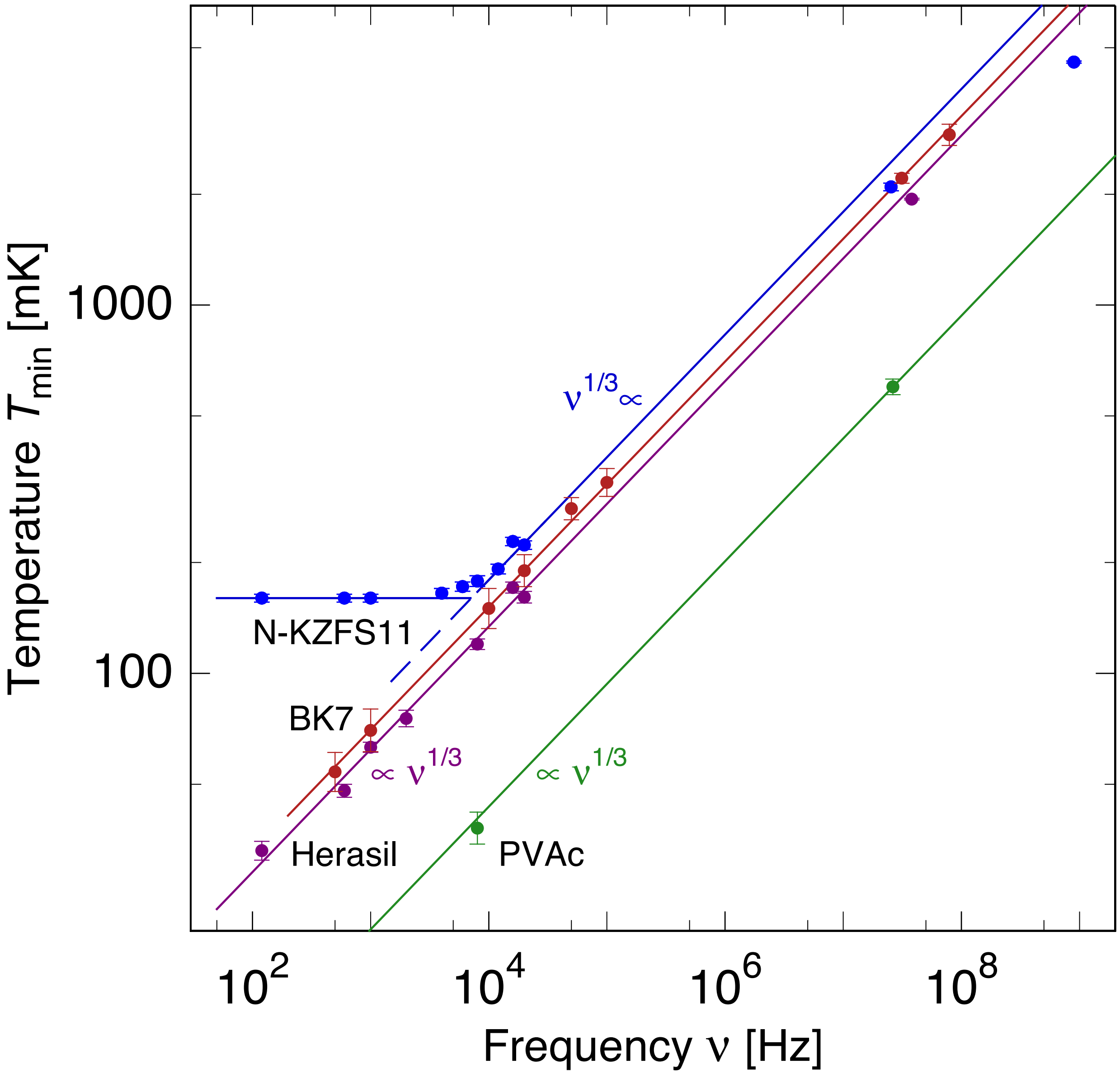

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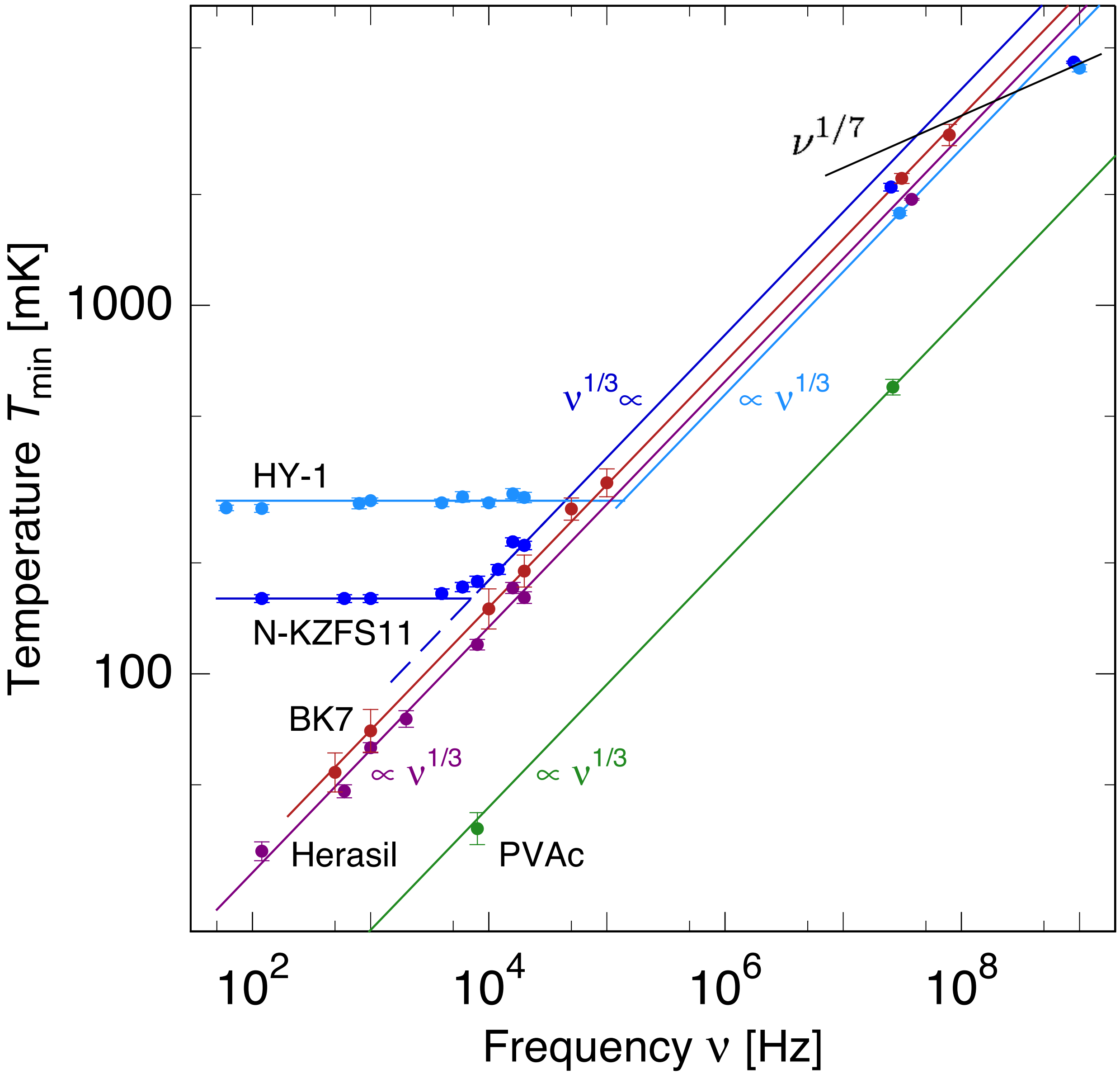

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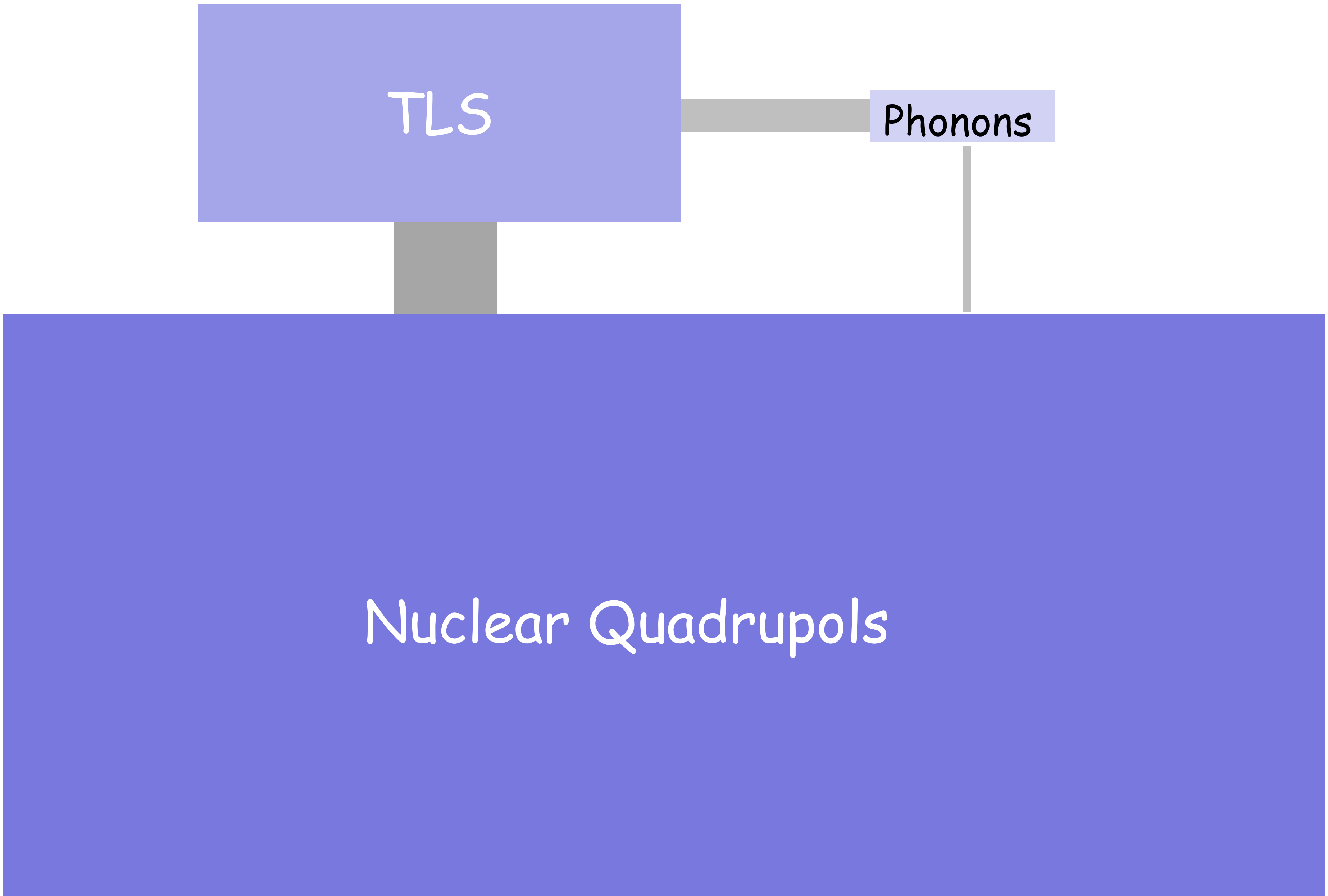
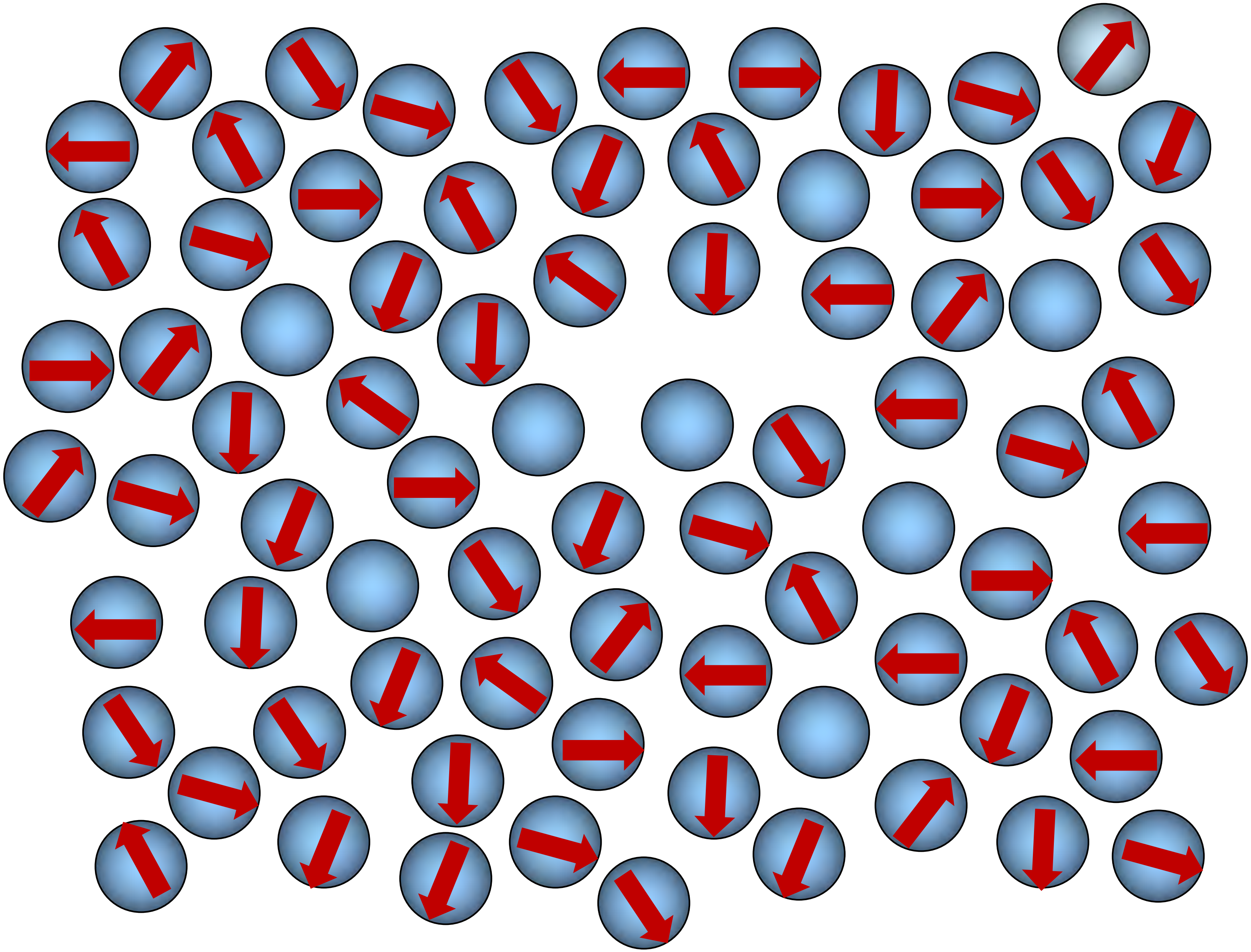
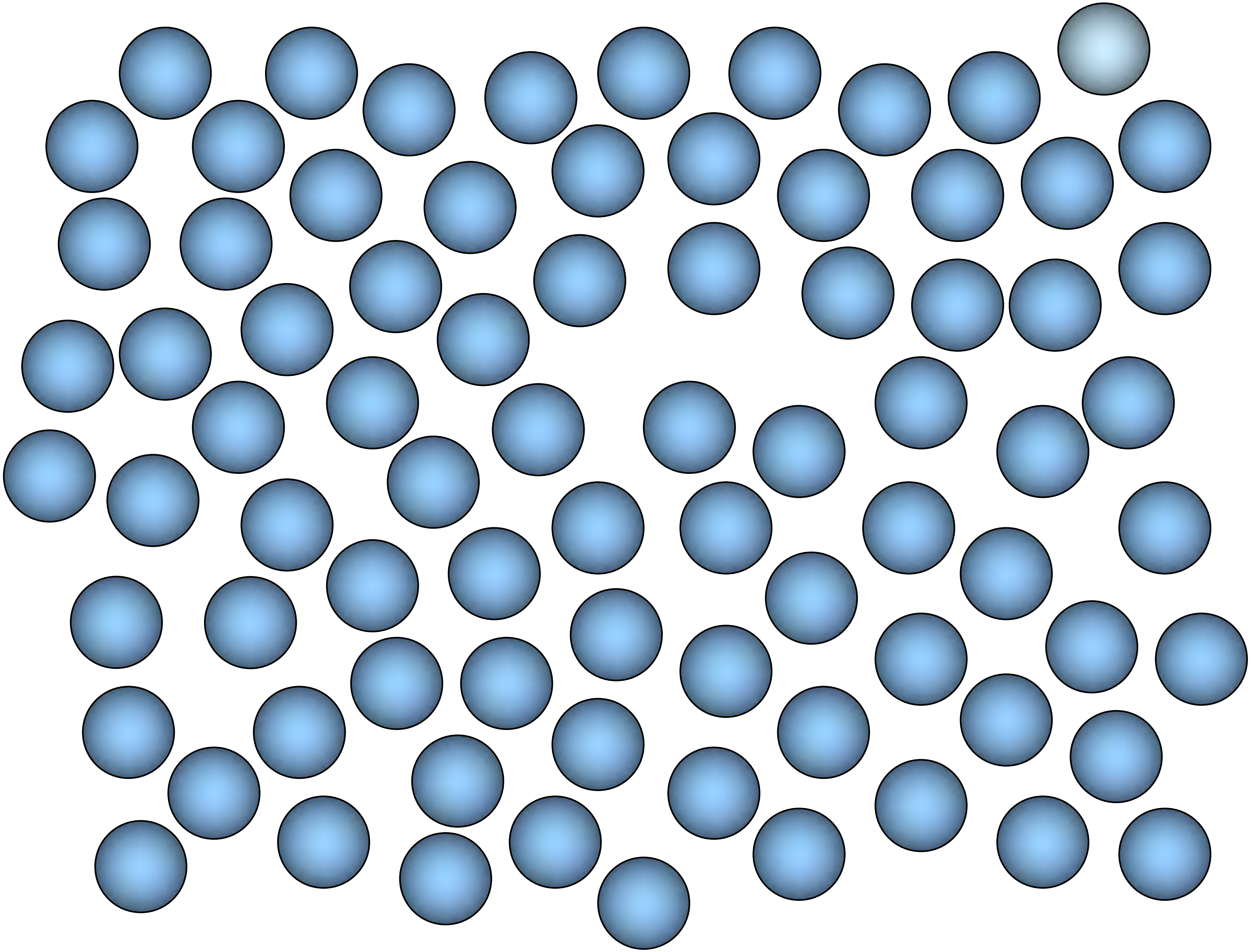
Relaxation Into Nuclear Spin Bath



Relaxation Into Nuclear Spin Bath



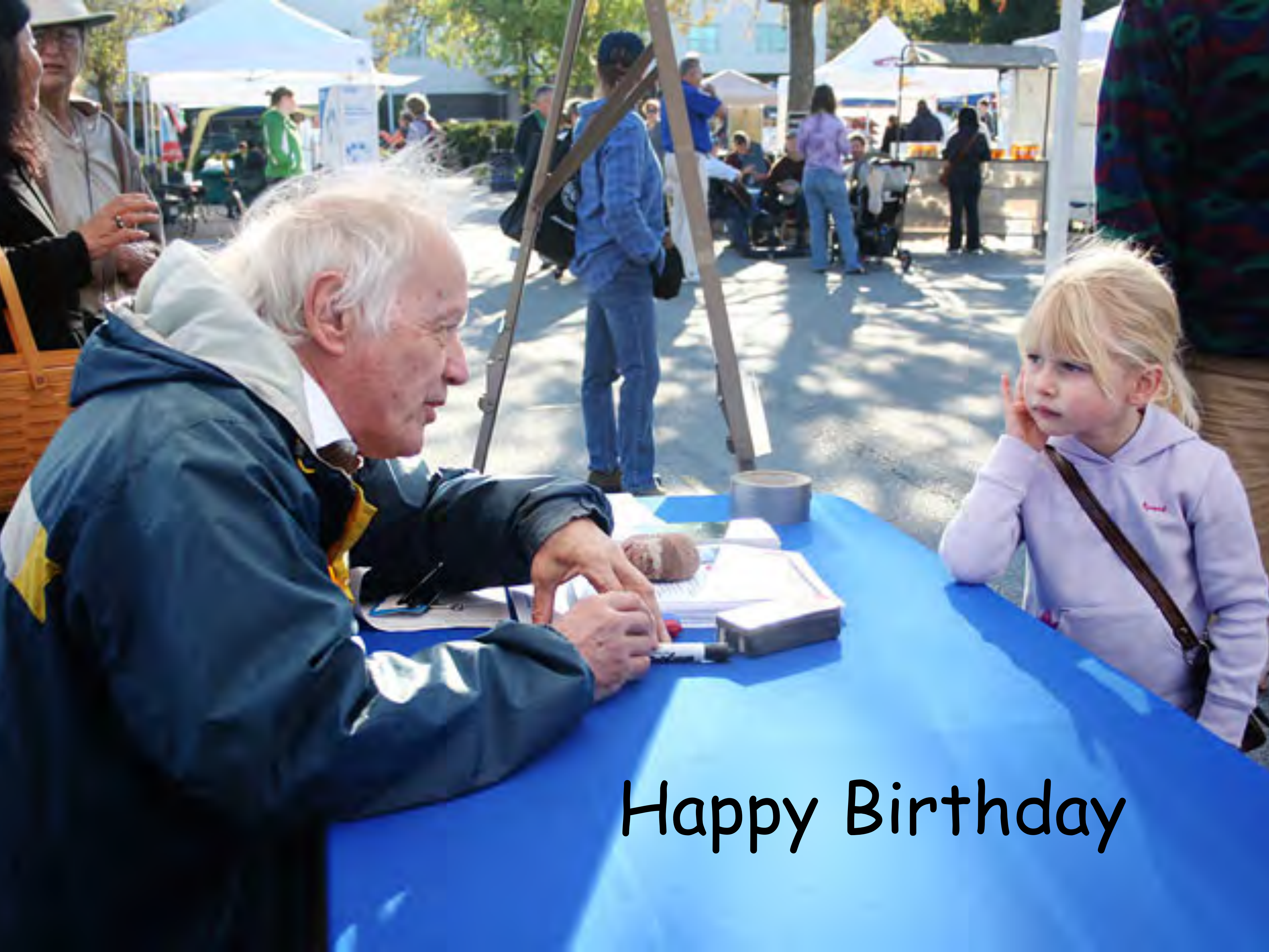
Atomic Tunneling Systems in Glasses



Summary

- Nuclear spins are important in glasses at low temperatures
- The universality of glasses breaks down at very low temperatures
- This allows for material dependent studies
- TLS model can be underpinned microscopically at least for some materials
- Still no microscopic explanation for the universality in glasses

There are still many fundamental questions regarding glasses to think about and work on


Happy Birthday