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

Catania 1999
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.
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Comments Cond. Mat. Phys.
Amorphous materials at low temperatures: why are they so similar?

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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$_2$

- **Vitreous silica**
  - Broad distribution of low-energy excitations

- **Quartz**

**Graph Details:**
- **Y-axis:** Specific Heat $C_V$ (μJ/gK)
- **X-axis:** Temperature $T$ (K)
- **Equation:** $T^3$

**References:**
Specific Heat of Amorphous and Crystalline SiO$_2$

R.C. Zeller, R.O. Pohl, 

excitations are localized

strong coupling to phonons
Universality of Glasses

\[ \text{Thermal conductivity of glasses within one order of magnitude} \]

Atomic Tunneling Systems in Glasses

P.W. Anderson et al., Philos. Mag. 25, 1 (1972)

energy splitting

\[ E = \sqrt{\Delta_0^2 + \Delta^2} \]

distribution function

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

tunnel splitting

\[ \Delta_0 = \frac{\hbar \Omega}{2} e^{-\lambda} \]
\[ \lambda = \frac{d}{2\hbar} \sqrt{2mV} \]

elastic, dielectric und thermal properities
Is there an unambiguous test for TLS in glasses?


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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.

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


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

Proof: Isotope Effect $H \leftrightarrow D$

**Glycerol**

\[
\begin{align*}
\text{hydrogen} & \quad I = 1/2, \quad \mu = 2.79 \mu_N, \quad Q = 0 \\
deuterium atom & \quad I = 1, \quad \mu = 0.86 \mu_N, \quad Q = 0.0029 b
\end{align*}
\]


\[C_3H_8O_3\quad C_3D_8O_3\]

\[t_{12} = 3.5 \mu s\]
Zero Magnetic Field

A. Würger, JLTP 137, 143 (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

\[ \nu_Q = 128 \text{ kHz} \]
\[ \nu_Q = 125 \text{ kHz (NMR)} \]

Beating in case of \textbf{d3} disappears faster

\[ \nu_Q = 160 \text{ kHz} \]
\[ \nu_Q = 158 \text{ kHz (NMR)} \]


Microscopic Modeling

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

M. Bazrafshan, PhD Thesis  2008


First measurement of microscopic properties of TLS in Glass
Can nuclear spins explain the magnetic field dependence of the dielectric susceptibility?
Coupling to Electric and Elastic Fields

**Resonant Processes**

\[ \hbar \omega \downarrow \quad E \uparrow \]

**Relaxational Processes**

\[ \delta \Delta = 2 \gamma e \]
\[ \delta \Delta = 2 p \cdot F \]

\( T < 1 \text{ K} \) one-phonon relaxation

\[ \tau_1 = A \left( \frac{E}{\Delta_0} \right)^2 \frac{1}{E^3} \tanh \left( \frac{E}{2k_B T} \right) \]
Dielectric Susceptibility in the STM

Real part after integration of tunnling parameter distribution
Different Frequencies

Relaxation Rate

$\tau^{-1}_{1\text{ph}} \propto E^3 \propto T^3 \quad \Rightarrow \quad T_{\text{min}} \propto f^{1/3}$  

one phonon

$\tau^{-1}_{2\text{ph}} \propto T^7 \quad \Rightarrow \quad T_{\text{min}} \propto f^{1/7}$  

two phonon

![Graph showing relaxation rates and temperatures](image-url)
### Samples

<table>
<thead>
<tr>
<th>Name</th>
<th>Type of Glass</th>
<th>Quadrupole Moment $Q$</th>
</tr>
</thead>
<tbody>
<tr>
<td>CVAc</td>
<td>Polymer glass</td>
<td>None</td>
</tr>
<tr>
<td>Herasil</td>
<td>Pure quartz glass</td>
<td>None</td>
</tr>
<tr>
<td>BK7</td>
<td>Multi-component glass</td>
<td>No large $Q$</td>
</tr>
<tr>
<td>N-KZFS11</td>
<td>Multi-component glass</td>
<td>$^{181}$Ta, $Q=3.3$ barn</td>
</tr>
<tr>
<td>HY-1</td>
<td>Multi-component glass</td>
<td>$^{165}$Ho, $Q=3.5$ barn</td>
</tr>
</tbody>
</table>
Dielectric Susceptibility of Standard Glasses

Herasil

Multi-component glass BK7

There is an additional observation that we would like to mention without going into a detailed discussion. Below 100 mK, the real and imaginary parts of the elastic and dielectric susceptibility exhibit pronounced non-linearities. The reason is that, with decreasing temperature, tunneling systems with smaller and smaller energy splitting become important. For these systems the assumption originally made in Sect. 9.1.2, namely, that changes caused by the external fields are small compared to the energy splitting in zero field, is no longer fulfilled. Fields of moderate strength already cause changes $\delta E$.

We do not discuss this interesting aspect further but refer the reader to the literature [443, 444].

Non-equilibrium Phenomena – Memory Effect

The temporarily application of a strong dc electric field across a capacitor with a glassy dielectric results in a sudden jump of the capacitance (which is proportional to the dielectric constant of the glass in this measurement) and a subsequent slow relaxation towards its starting value [445, 446, 447]. As an example we show in Fig. 9.43a the time evolution of the capacitance of a capacitor filled with a-SiO$_x$ after the sudden application of a large dc electric field at 50 mK. The slow relaxation observed in this experiment is logarithmic in the time elapsed since the field was applied.

Another interesting observation can be made when slowly sweeping the dc field after the sample has been kept in a constant field for a long time. In this case the capacitance shows a minimum at the field at which the sample was kept prior the sweep experiment. Fig. 9.43b shows the response to such a sweep of a capacitor filled with photoresist (an amorphous polymer) which initially had been cooled down without an electric field and then kept at a field of $F = 5$ MV/m for about 2 hours. Clearly, a minimum of the capacitance was found at zero field, at which the sample was kept for a long time.
Frequency Dependence of Minimum in DK

One phonon process:

\[ T_{\text{min}} \propto \nu^{1/3} \]

Two phonon process:

\[ T_{\text{min}} \propto \nu^{1/7} \]
Dielectric Susceptibility of Glasses With Nuclear Quadrupols

Multi-component glass containing Tatalum

Dielectric Susceptibility of Glasses With Nuclear Quadrupols

Multi-component glass containing Tatalum

Multi-component glass containing Holmium

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

One phonon process:

\[ T_{min} \propto \nu^{1/3} \]

Two phonon process:

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Relaxation Into Nuclear Spin Bath

TLS

Phonons

Nuclear Quadrupols
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