Title: Glass: The Cinderella Problem of Condensed Matter Physics

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Abstract: Amorphous materials (glasses) probably constitute >90% of the solid matter surrounding us in everyday life,yet traditional textbooks of condensed matter physics devote virtually no space to them.Crudely speaking,the puzzles in the behavior of glasses can be divided into three major areas:the glass transition itself,the characteristic long-term memory effects and the near-equilibrium thermal,dielectric and transport properties;this talk focusses entirely on the third area.Over the last 40 years it has become apparent that the thermal and transport properties are not only qualitatively universal between glasses with totally different chemistry and microstructure,but in some cases possess a truly mind-boggling degree of _quantitative_ universality.In this talk I will describe the salient experimental data,review the established ("tunnelling two-level system") model commonly used to interpret them,and introduce a rather different scenario (developed in collaboration with D.C.Vural) which holds out some prospect of explaining the universalities in a natural way.

GLASS:
THE CINDERELLA PROBLEM OF CONDENSED-MATTER PHYSICS

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University of Illinois at
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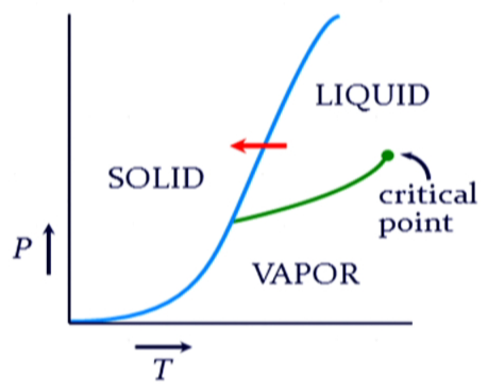
GLASS

Typical "textbook" phase diagram (H_2O):



Liquid phase:

a) (Time-averaged) density spatially uniform
b) Cannot sustain shear

Crystalline solid:

a) If we define

$\rho_k \equiv \Omega^{-1}\int d\underset{\sim}{r}\, e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} \langle \rho(\underset{\sim}{r})\rangle$, (time-averaged density)

then for some $\underset{\sim}{K} \neq 0$ $\rho_K \neq 0$

b) Shear modulus nonzero

UW 8.2

General belief:

a) In thermodynamic limit, GS (groundstate) of any element or mixture of elements is either a crystalline solid or (exceptionally: He!) a liquid (↑: quasicrystals!)
b) On sufficiently slow cooling at constant P, this GS will be obtained

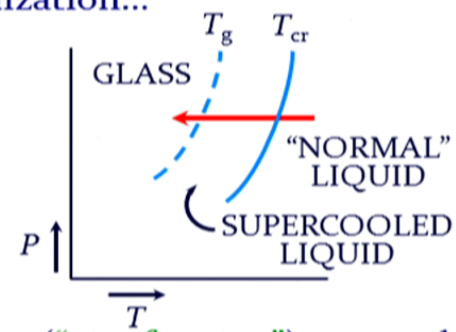
But:

a) What is "sufficiently slow"?
b) What happens if it is not?

Intuitively, expect fast crystallization for *e.g.* single non-gas elements (?: mixtures of rare gases?), much slower for *e.g.* large biomolecules.

UW 8.3

If no crystallization…



Glass transition ("vitrification"): not a phase transition, but a dramatic change of kinetic coefficients, *e.g.* huge change in viscosity (up to 17 orders of magnitude).

"T_g" a matter of convention and somewhat cooling-rate-dependent: usual convention point at which $\eta \sim 10^{12}$ Pa□s (1 Pa□s ≡ 1 kg/m·sec: η of H_2O at RT ~ 10^{-3} Pa□s) T_g always < T_s, typically ~ 0.7 T_s.

UW 8.4

The glass ("amorphous solid") phase defining characteristics:

a) $\rho_k \sim 1/\Omega^d$ (d < 0) for all $\underset{\sim}{k} \neq 0$ *i.e.* no crystalline order

b) However, $\lim_{T\to\infty} T^{-1}\int_0^T \langle \rho(\underset{\sim}{r}0)\rho(\underset{\sim}{r}t)\rangle \mathrm{d}t - \rho_o^2 \neq 0$ in general (*i.e.* not liquid) (ρ_o: mean density)

c) Nonzero shear modulus over "experimental" time scales.

Are glasses just "extremely viscous" liquids?
cf. the cathedral-window problem*

*Zanotto and Gupta, Am. J. Phys. **67**, 260 (1999).

UW 8.5

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UW 8.5

Types of glasses:

- Silicate (SiO_2, GeO_2)
- Borosilicate (B_2O_3,...)
- Lead-crystal (PbO)
- High-polymer
- Electrolyte solution
- ...

May contain electric/magnetic dipole moments.
May show ferromagnetism/spin glass/normal-metallic (*e.g.* PdCuSi)/even superconductivity (*e.g.* NbTi)

For nonmagnetic, nonmetallic glasses, 3 main classes of problem:

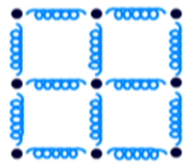
1) The glass transition
2) Aging and relaxation effects

} explore whole phase space

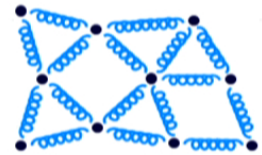
3)* Thermal and ultrasonic (and dielectric) properties — explore region close to local minimum

UW 8.6

A "minimal" picture of the thermal and ultrasonic properties of glasses: the "disordered-harmonic" model.



crystalline solid



glass

In each case,

$$\hat{H} = \sum_i p_i^2/2m_i + \tfrac{1}{2}\sum_{i,j} V(\underset{\sim}{r}_i - \underset{\sim}{r}_j)$$

$$\cong \sum_i p_i^2/2m_i + \tfrac{1}{2}\sum_{i,j,\alpha,\beta} V_{ij\alpha\beta}u_{\alpha i}u_{\beta j}$$

$$u_{\alpha i} \equiv \underset{\sim}{r}_{\alpha i} - \underset{\sim}{R}^{(0)}_{\alpha i}$$

$\Rightarrow$ can be exactly diagonalized:

$$\hat{H} = \sum_i \hbar\omega_i a_i^{+} a_i$$

UW 8.7

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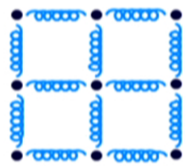

crystalline solid

glass

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⇒ can be exactly diagonalized:

$$\hat{H} = \sum_i \hbar\omega_i a_i^+ a_i$$

UW 8.7

Consequences:

1. No nonlinear effects (response of SHO's strictly linear)
2. $c_v(T) = \int_0^\infty \rho(\omega) \frac{dn(\omega)}{dT} d\omega$ — Bose distribution; $\rho(\omega) = \sum \delta(\omega - \omega_i)$
3. For $\omega \to 0$, only contribution is from long-wavelenth longitudinal (l) and transverse (t) modes of elastic continuum $\Rightarrow \quad \rho(\omega) \propto \omega^2$ (just as in crystalline case)
4. Expect also higher-frequency localized modes
5. In approximation that under both compressional and shear stress $\delta \underset{\sim}{r}_{ij} \propto \left| \underset{\sim}{R}_i^o - \underset{\sim}{R}_j^o \right|$, predict $c_t / c_l = \frac{1}{\sqrt{3}}$. (Lord Rayleigh 1886?)

uw 8.8

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UW 8.8

Of these predictions, (4) seems consistent both with intermediate-temperature behaviour of specific heat ("boson peak" in c_v-c_v^{Debye}) and with inelastic neutron scattering. Also (5) verified within <10%. However:

(2) + (3) → in limit $T \to 0$, $c_v \propto T^3$ (Debye law)

Not true! (Pohl, 1970): for almost all amorphous solids below ~1 K,

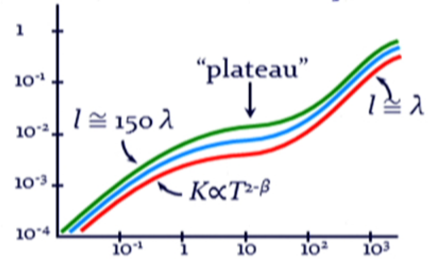
$$c_v(T) \propto T^{1+\alpha},\ \alpha \sim 0.2\text{-}0.3$$

(↑: could almost certainly fit equally well to $c_v(T) \propto T \ln(T_o/T)$)

⇒ extra low-energy DOF's besides long-wavelength phonons ↳degrees of freedom

UW 8.9

Universality of thermal conductivity:



$\propto \frac{\theta_D^2}{\bar{c}}$ "Debye" T

Plot of $\kappa/\bar{\kappa}$ vs. T/θ_D looks even more universal*, in particular both low-T and high-T data collapse on to single curve.

It seems certain that at least for $T < 1$ K thermal transport is entirely or mainly by phonons‡. If so, data indicate that phonon (ultrasound) mean free path for $\hbar\omega \sim k_B T$ is $\approx 150\,\lambda$, or equivalently $Q_{hf}^{-1} \approx 3 \times 10^{-4}$.

This is consistent with direct measurements of ultrasound absorption and velocity.

*Anderson and Freeman, Phys. Rev. B **34**, 5684 (1986).
‡Zaitlin and Anderson, Phys. Rev. B **12**, 4475 (1975).

UW 8.10

Tunnelling 2-level system (TTLS) model

In "site" basis, 2LS Hamiltonian is $\begin{pmatrix} \varepsilon & \Delta \\ \Delta & -\varepsilon \end{pmatrix}$ so $E = \pm\sqrt{\varepsilon^2 + \Delta^2}$.

What is DOS of 2LS? $\rho(E) = \int \mathrm{d}E \int \mathrm{d}\Delta\; P(\varepsilon,\Delta)\, \delta\left(E - \sqrt{\varepsilon^2+\Delta^2}\right)$,

(DOS = density of states)

so need $P(\varepsilon,\Delta)$.

Ansatz:

a) $P(\varepsilon,\Delta) = f(\varepsilon)g(\Delta)$ (*i.e.* no correlation between ε and Δ)

b) $f(\varepsilon) \approx$ const.

c) Since $\Delta \sim \omega_o \exp\text{-}\lambda$, if we assume $\rho(\lambda)$ = const. (big if!), then $g(\Delta) \sim \Delta^{-1}$, so

(λ: WKB experiment $\sim \int \sqrt{2mV(r)}\,\mathrm{d}r/\hbar$)

$$p(E) - \int \mathrm{d}\varepsilon \int \tfrac{\mathrm{d}\Delta}{\Delta}\, \delta\left(E - \sqrt{\varepsilon^2+\Delta^2}\right) = \text{const.}$$

(standard notation: $\rho(E) \rightarrow \bar{P}$)

($\bar{P}$: per unit volume)

UW 8.11

If so, then $c_v(T) = \int_0^\infty \mathrm{d}E\,\rho(E)\,\mathrm{sech}^2\, E/2T \propto T$
approximately as experiment.

ULTRASOUND ATTENUATION AND VELOCITY*

Generally speaking, in "canonical" (energy) basis, TLS couple to strain field through both $\hat{\sigma}_x$ and $\hat{\sigma}_z \Rightarrow$ absorption of ultrasound (phonons) and spontaneous phonon emission (relaxation), with some "typical" relaxation time τ. (= $f(E)$, hence $f(T)$)

*J. Jäckle, Z. Phys. A **257**, 212 (1972)

UW 8.12

2 major regimes, distinguished by value of $\omega\tau$:

A. **"Resonance" regime ($\omega\tau \gg 1$)** (σ_x-coupling)

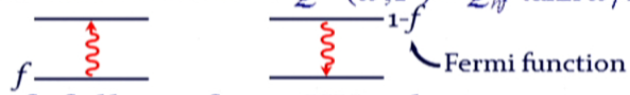
Absorption of US by direct excitation of TLS:

$\Rightarrow$ at $T = 0$, $Q^{-1}(\omega)$ independent of ω and

mean-square coupling to strain

given by $Q^{-1}(0) = \pi \dfrac{\overline{\gamma}^2 \overline{P}}{\rho c^2} \equiv Q_{hf}^{-1}$.

At finite T, need to consider both absorption and stimulated emission: $Q^{-1}(\omega, T) = Q_{hf}^{-1} \tanh \omega / 2T$



Velocity shift follows from KK relation:

$$\frac{\Delta c}{c_o} \propto \int_0^\infty \frac{Q^{-1}(\omega', T) \tanh \omega'/2T}{\omega - \omega'} = Q_{hf}^{-1} \ln\left(\frac{T}{T_o}\right)$$

(usually most accurate way of measuring Q_{hf}^{-1})

UW 8.13

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UW 8.13

In addition, TTLS model makes confident qualitative predictions on a variety of nonlinear phenomena (saturation, echoes, hole-burning...). All seen experimentally! (And can be fitted quantitatively with appropriate choice of fitting factors.)

So...what could possible be wrong with TTLS?

1. In most glassy systems, no microscopic picture of nature of TLS. (Exception: $KBr_x(CN)_{1-x}$)
2. No explanation of higher-T phenomena, *e.g.* plateau in κ.
3. (In simplest form) no account of inter-TLS interaction.
4. Cannot explain universality of Q_{hf}^{-1} except by extraordinary coincidence:

$$Q_{hf}^{-1} = \frac{\pi \bar{\gamma}^2 \bar{P}}{\rho c^2} \Bigg\}$$ in TLS model, 4 independent quantities each fluctuating by ~ 5-10

= (3±2) × 10^{-4} for (essentially) all known glasses.

UW 8.15

An Alternative Model of the Low-Temperature Thermal and Acoustic Properties of Glasses

(D.C. Vural and A.J.L., J. Noncrystalline Solids 357, 3528 (2011))

The basic idea:

start with phonons and arbitrary other DOF's, subject only to requirement that spectrum is not harmonic.

Effects encapsulated in

(a) Hamiltonian $\hat{H}(e_{ij}=0) \equiv \hat{H}_0$

(b) stress tensor $\hat{T}_{ij} \equiv (\partial \hat{H}/\partial e_{ij})_{e_{ij}=0}$

so

$$\hat{H} = \hat{H}_{ph} + \hat{H}_0 + \sum_{ij} \hat{e}_{ij} \hat{T}_{ij}$$

Coupling to phonons ⇒ induced stress-stress interaction

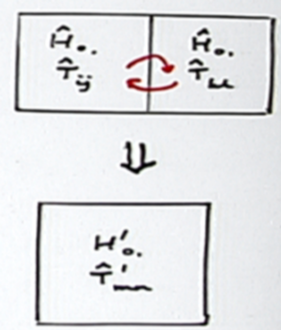
$$\hat{H} \to \hat{H}_0' + \sum_{ijkl} \iint \frac{d\mathbf{r}\, d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|^3} \Lambda_{ijkl} \hat{T}_{ij}(\mathbf{r}) \cdot \hat{T}_{kl}(\mathbf{r}')$$

Basic object of study: stress-stress correlation function

$$\mathrm{Im}\, \chi_{ij:kl}(\omega) \equiv \sum_m p_m \chi^{(m)}_{ij:kl}(\omega),$$

$$\chi^{(m)}_{ij:kl}(\omega) \equiv \sum_n \langle m|\hat{T}_{ij}|n\rangle\langle n|\hat{T}_{kl}|m\rangle \, \delta(\epsilon_n - \epsilon_m - \hbar\omega)$$

$$Q^{-1}(\omega) = \text{const.} \cdot \mathrm{Im}\, \chi(\omega)$$



Qualitative intuition (language of TTLS model – special case!): $(\hat{T}_{ij} \to \gamma)$

$$Q^{-1} \sim \frac{\gamma^2 \bar{P}}{\rho c^2}$$

what is $\bar{P}$? if assume interaction terms dominate,

induced interaction $\propto g/r^3$, $g \sim n\gamma^2/\rho c^2$

on dimensional grounds, $\bar{P} \propto g^{-1}$

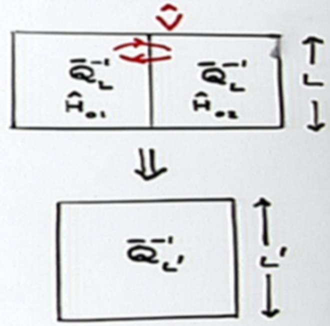
$\Rightarrow \quad \bar{P} \sim \rho c^2/n\gamma^2$

$\Rightarrow \quad Q^{-1} \sim n^{-1}$ and is universal!

Problem: in our case $n = 3$, so prima facie $Q^{-1} \sim 1$!

(expt: $Q^{-1} \sim 3 \times 10^{-4}$)

$$\hat{H} = \hat{H}_0 + \hat{V}$$
$$\hat{H}_0 = \hat{H}_{01} + \hat{H}_{02}$$
$(\hat{H}_0, \hat{V}$ uncorrelated)



$$\mathrm{Tr}\,\hat{H}^2 = \mathrm{Tr}\,\hat{H}_0^2 + \mathrm{Tr}\,\hat{V}^2$$

$$\mathrm{Tr}\,\hat{H}_0^2 = N_s^2 U_0^2, \quad \mathrm{Tr}\,\hat{H}^2 = N_s^2 U^2$$

$$\mathrm{Tr}\,\hat{V}^2 = CK N_s^2 U_0^2 (\bar{Q}_L^{-1})^2$$
$$\cong CK N_s^2 U^2 (\bar{Q}_{L'}^{-1})^2$$

$C \equiv$ geometrical factor $(= 9/16\pi^2)$

$K \equiv 8/3\,[-3 + 4p + 16q(q + p + qp - 1)]$

$p \equiv \ln \chi_\ell / \ln \chi_t \cong 2.6$
$q \equiv 1 - c_t^2/c_\ell^2 \cong 0.6$
} quasi-universal

$\Rightarrow \quad K \approx 122$

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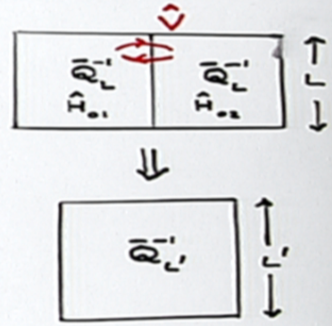

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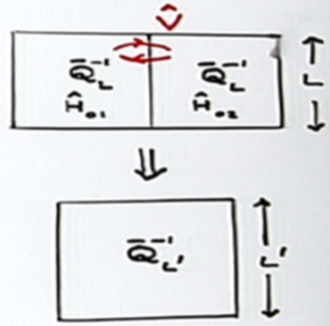

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$\Rightarrow K \approx 122$

Output of single step, combining 8 blocks of side L to make one of side 2L: [RL 3]

$$\bar{Q}^{-1}_{(2L)} = \left[\frac{1}{(\bar{Q}^{-1}_L)^2} + K_0\right]^{-1/2} \qquad K_0 \sim 150$$

Iterate procedure to experimental length scale R:

$$\bar{Q}^{-1}(R) = \left[\bar{Q}_L^2 + K_0 \log_2(R/L)\right]^{-1/2}$$

Note predicts $\bar{Q}^{-1} \to 0$ for $R \to \infty$! When $R \sim$ exptl. length scale,

$$\bar{Q}^{-1} \sim 0.015$$

Note $\bar{Q}^{-1}$ is average ultrasound absorption over large frequency range, hence only $= \bar{Q}^{-1}_{exp}$ if $Q^{-1}(\omega) =$ const. Is this likely to be so?

Clue: KK rel? $\Rightarrow$ static compressibility $\propto \int_0^\infty \frac{Q^{-1}(\omega)}{\omega} d\omega$ $\Rightarrow$ diverges if $Q^{-1}(\omega) =$ ind. of ω.

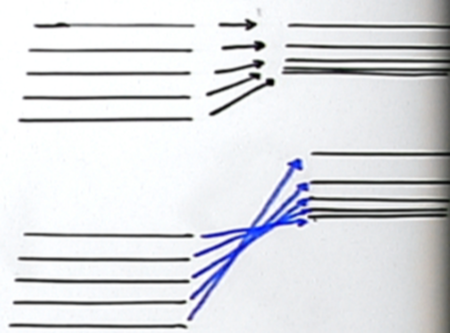
To avoid divergence, $Q^{-1}(\omega)$ needs to decrease with ω at least as fast as $(\ln \omega)^{-(1+\epsilon)}$, $\epsilon > 0$. Does it?

What is result of treating $\hat{V}$ by 2nd order perturbation theory?

Ans.: low-energy DOS increases by factor $(\ln \epsilon)^{-1}$!

?

2nd-order perturbation theory (cont): Vural conjecture



Formal result:

$$R(\epsilon) = (1 + \partial\Delta/\partial\epsilon)^{-1}$$
$$= (1 - cKQ_0^{-2}\ln(U_0/\epsilon))^{-1}$$

which is negative for small ϵ (unphysical).

Conjecture: take

$$R(\epsilon) = |(1 + \partial\Delta/\partial\epsilon)|^{-1}$$
$$= [cKQ_0^{-2}\ln(U_0/\epsilon) - 1]^{-1}$$

then on iteration get

$$Q^{-1}(\omega) = \frac{Q_0^{-1}}{K_0 Q_0^{-2}(\ln_2 R/L)(\ln U_0/\omega) - 1} \sim \frac{1}{\ln(U_0/\omega)}$$

but, doesn't agree with result for $\overline{Q^{-1}}(\omega)$.

So, conjecture:

$$Q^{-1}(\omega) = \frac{1}{Q_0 + A\ln(U_0/\omega)} \qquad (*)$$

where A fitted from consistency with result for $\overline{Q_0^{-1}}$. This gives, for $\omega \sim$ expt. value, ($U_0 \sim \hbar c/L_0$, $L_0 \sim 50$ Å)

$$Q^{-1}(1\,\text{MHz}) \approx 2.7 \times 10^{-4}$$

Appears to fit ultrasonic data reasonably well, and predicts

$$K \sim T^2 \ln(U_0/T)$$

(easily mistaken for $T^{2-\beta}$, $\beta \sim 0.05 - 0.2$.)

RL 3

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Note predicts $\bar{Q}^{-1} \to 0$ for $R \to \infty$! When $R \sim$ expt. length scale,

$$\bar{Q}^{-1} \sim 0.015$$

Note $\bar{Q}^{-1}$ is average ultrasound absorption over large frequency range, hence only $= \bar{Q}^{-1}_{exp}$ if $Q^{-1}(\omega) =$ const. Is this likely to be so?

Clue: KK rel^n $\Rightarrow$ static compressibility $\propto \int_0^\infty \frac{Q^{-1}(\omega)}{\omega} d\omega$ $\Rightarrow$ diverges if $Q^{-1}(\omega) =$ ind. of ω.

To avoid divergence, $Q^{-1}(\omega)$ needs to decrease with ω at least as fast as $(\ln \omega)^{-(1+\epsilon)}$, $\epsilon > 0$. Does it?

What is result of treating $\hat{V}$ by 2nd order perturbation theory?

Ans.: low-energy DOS increases by factor $(\ln \epsilon)^{-1}$!

?

If above arguments are right, observed value of $\overline{Q^{-1}}$, namely $\sim 3 \times 10^{-4}$, is the maximum obtainable. It then follows that we can set an **absolute lower limit** on K for **any** material (including crystalline).

Some further questions:

1) Can we make the arguments more rigorous?
   (? "Efros–Shklovskii" approach?)
2) US absorption in the "relaxational" regime
   ($\omega\tau \ll 1$)
3) nonlinear phenomena (saturation, echoes, hole-burning ...)
4) plateau in K (+ "boson peak" in c_v)
5) high-temperature behavior of K (quasi-universal: indicates $\overline{Q^{-1}}(\omega) \to \sim 1$ for $\omega \gg \omega_0 \sim c/L$
   ("Ising–Heisenberg crossover"?)

SLOGAN FOR THE 21ST CENTURY:

GLASSES ARE THE NORM.

CRYSTALS THE ANOMALY!