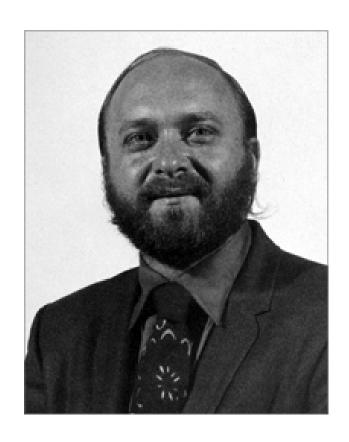
# Seeking simplicity in complexity: A physicist's view of vulcanized media

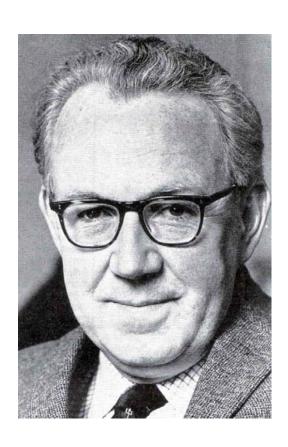
PAUL M. GOLDBART
School of Physics
Georgia Institute of Technology



**Leo Kadanoff** (1937-2015)

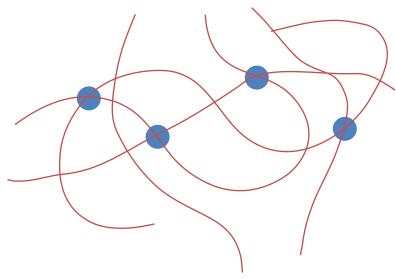


Ken Wilson (1936-2013)



Sam Edwards (1928-2015)

# **VULCANIZED MATTER**



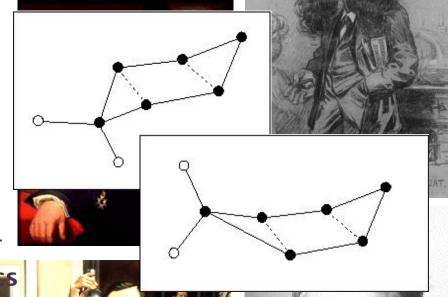
- what is it?
- what's unusual about it?
- why is it *complex*?

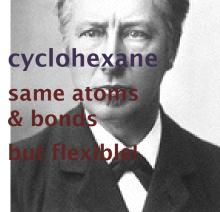
# LET'S BEGIN WITH MOLECULAR MATTER

- o structure of matter ~ mid 1800's
  - Berzelius, Kekulé, Pasteur, van 't Hoff,...
  - chemical reactions, optical activity
- o emerging picture
  - molecules = 3D patterns of chemically bonded atoms
  - structural- & stereo-isomers...

o classification, but not dynamics

- until quantum theory





# JUST ONE OF VAN 'T HOFF'S CRITICS...

"...there is an overgrowth of the weed of the seemingly learned and ingenious but in reality trivial and stupefying...

[which is] being dragged out by pseudoscientists from the junk-room which harbors such failings of the human mind, and...

[is being] dressed up in modern fashion and rouged freshly like a whore whom one tries to smuggle into good society where she does not belong."

"Whoever considers this apprehension to be exaggerated should read, if he can manage it, the recently published pamphlet, "The arrangement of atoms in space"... which teems with fantastic trifles."

Hermann Kolbe (1818-1884)

"The modern chemical theory has two weak points. It says nothing either about the relative position or the motion of the atoms within the molecule."

Jacobus Henricus van't Hoff (1852-1911), Chemistry Nobel Laureate (1901), in "The arrangement of atoms in space"

# NOW FOR MACROMOLECULAR MATTER

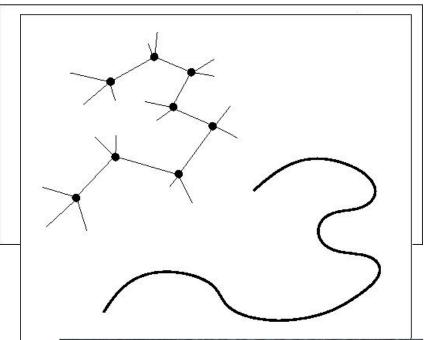
- o much longer molecules (eg PE)
- o amplified flexibility, random coils
- o but do they exist?

"Dear colleague, drop the idea... large molecules... with a molecular weight higher than 5000 do not exist. Purify your products... they will crystallize & prove to be low molecular compounds!"

Heinrich Wieland Chemistry Nobel Laureate (1927)

"colleagues were very skeptical [asking] why I was neglecting [low molecular chemistry for] *Schmierenchemie* or grease chemistry."

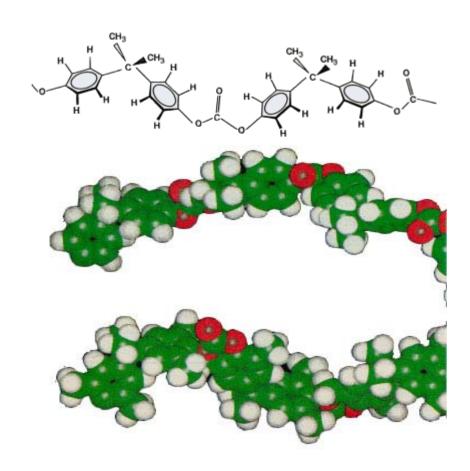
Hermann Staudinger, autobiography (1961) Chemistry Nobel Laureate (1953)





# SOME SCALES FOR A TYPICAL MACROMOLECULE

- monomers repeated <u>extraordinarily many</u> times
  - ~ 25,000 carbon atoms
  - ~ 75,000 atoms in all
  - ~ 5 µm along backbone
  - $\sim 0.1~\mu m$  across coil bending length  $\sim 1~nm$  length/dia.  $\sim 6{,}000$  25 m mouse cable



# as van 't Hoff & Staudinger surely understood...

"All truth passes through three stages: First, it is ridiculed; Second, it is violently opposed; and Third, it is accepted as self-evident."

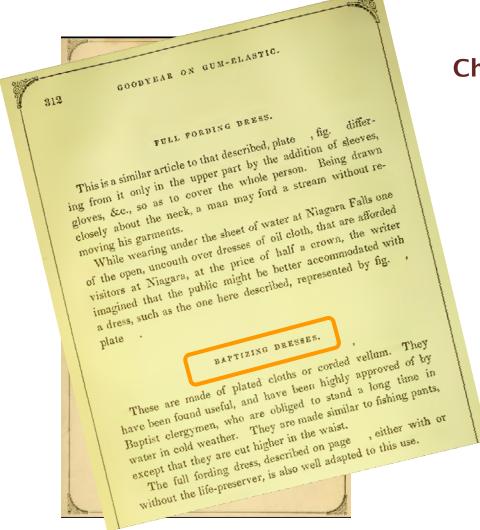
A. Schopenhauer (1788-1860)

# **MACROMOLECULAR MATTER**

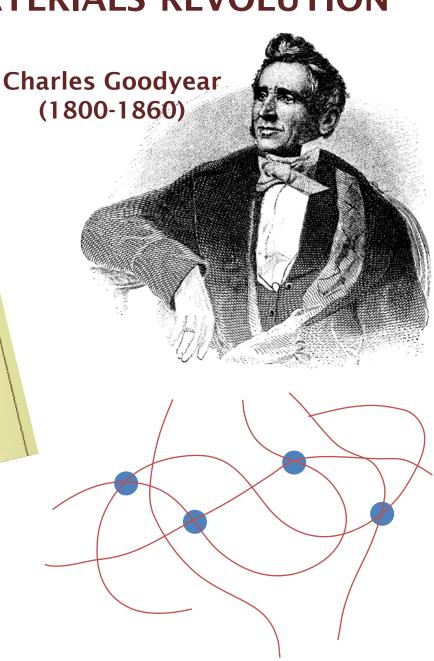
- e.g. natural rubber (polymerized isoprene), spider silk, polyethylene, chewing gum, plastics, resins...
- many biological & synthetic forms



**VULCANIZATION: A MATERIALS REVOLUTION** 

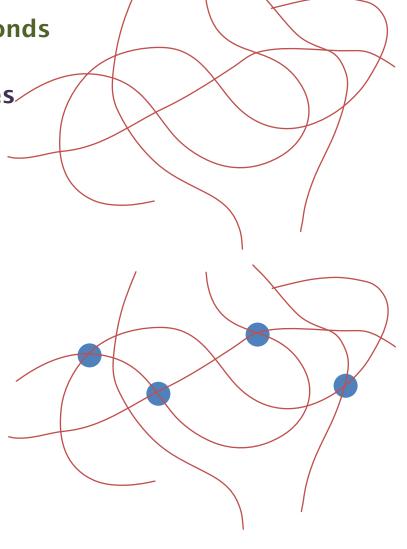


345 pages of commercial applications within 14 years of the discovery



# WHAT'S UNUSUAL ABOUT VULCANIZED MACROMOLECULAR

- randomly add permanent chemical bonds
  - into melts or solutions of macromolecules or small molecules
- what emerges ?
  - random space-filling network, thermally fluctuating
  - o random (but equilibrium!) solid
  - o similar to glass?
- architectural complexity
  - annealed but also quenched freedoms



vulcanization (vulcan, volcano,...)Hayward (1838), Goodyear (1839)

#### SCALES FOR VULCANIZED MACROMOLECULAR MATTER

- elastic moduli
  - o crystalline solids
    - bulk or shear

$$\sim {\rm Ry}/a_{\rm Bohr}^3 \sim m^4 e^{10}/h^8$$

- experimental values

 $\sim 10^{12} \mathrm{erg/cm}^3$ 

- o rubber
  - bulk: comparable
  - shear: ~100,000 times smaller
- reversible extensibility
  - o crystal: ~1%
  - rubber: ~700%

- o e.g. 1% extension of 1mm wire
  - **steel** ~ 350 lbs
  - rubber ~ fractions of an ounce

# GOAL: TO UNDERSTAND...

o emergent collective properties characteristic of this random solid

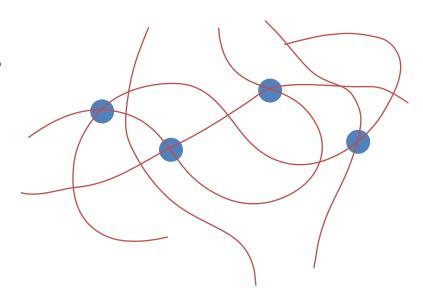
- o what properties ?
  - structure
  - elasticity
  - heterogeneity

emergent: not meaningful for an isolated polymer

- applications: blends, liquid crystalline elastomers (not today)
- o how can we capture these theoretically?
- o is there any universality to them?

# **OUTLINE**

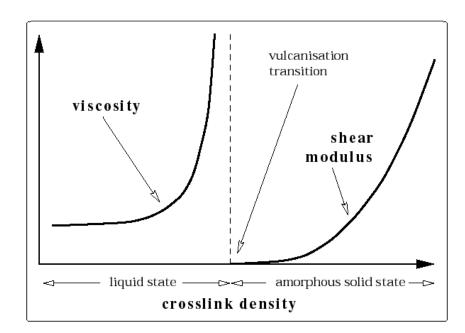
- o matter: molecular, macromolecular, vulcanized
- o the random solid state & its emergent collective properties
- o statistical mechanics, done statistically
- o detecting localization
- o order parameters & field theories
- o encoded emergent structure
- o encoded emergent elasticity
- o fluctuations & criticality
- o some remarks

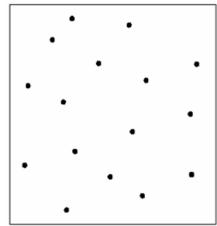


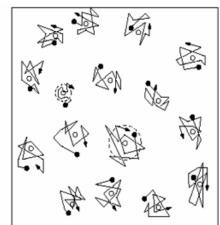
# SUFFICIENT VULCANIZATION TRIGGERS A RANDOM EQ'M SOLID

- first, add bonds...
- macro
  - o liquid to solid
  - o retains macro homogeneity

- micro
  - localization
  - o transl invariance spont broken
  - o but inhomogeneously
- can we understand...
  - o transition?
  - o implications?







## **DETECTING LOCALIZATION OF A PARTICLE**

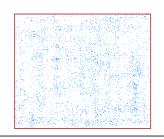
- begin with one particle
  - o position R
  - o choose wave vector k
  - $\circ$  equilibrium / thermal average  $\langle \cdots 
    angle$ 
    - if delocalized

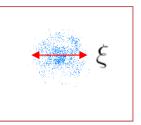
$$\langle \exp i\mathbf{k}\cdot\mathbf{R}\rangle = 0$$

- if localized

$$\langle \exp i\mathbf{k} \cdot \mathbf{R} \rangle \approx \exp i\mathbf{k} \cdot \langle \mathbf{R} \rangle \exp(-k^2 \xi^2/2)$$

random mean position





random r.m.s. displacement (localization length)  $\xi$ 

prob distrib of p'cle position

Aspen | July 2017

# **DETECTING THE RANDOM SOLID**

- collection of particles
  - o labels j = 1,...,J, positions  $\mathbf{R}_{j}$
  - $\circ$  averages: equilibrium  $\wedge$  , QRV  $[\Lambda]$
- a randomly localized particle

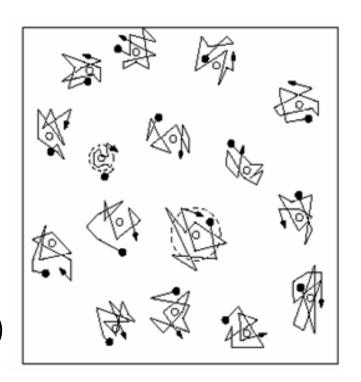
$$\langle \exp i\mathbf{k} \cdot \mathbf{R} \rangle \approx \exp i\mathbf{k} \cdot \langle \mathbf{R} \rangle \exp(-k^2 \xi^2/2)$$

• try 
$$\Omega(\mathbf{k}) = \left[\frac{1}{J}\sum_{j=1}^{J}\langle \mathrm{e}^{i\mathbf{k}\cdot\mathbf{R}_{j}}
angle
ight]$$

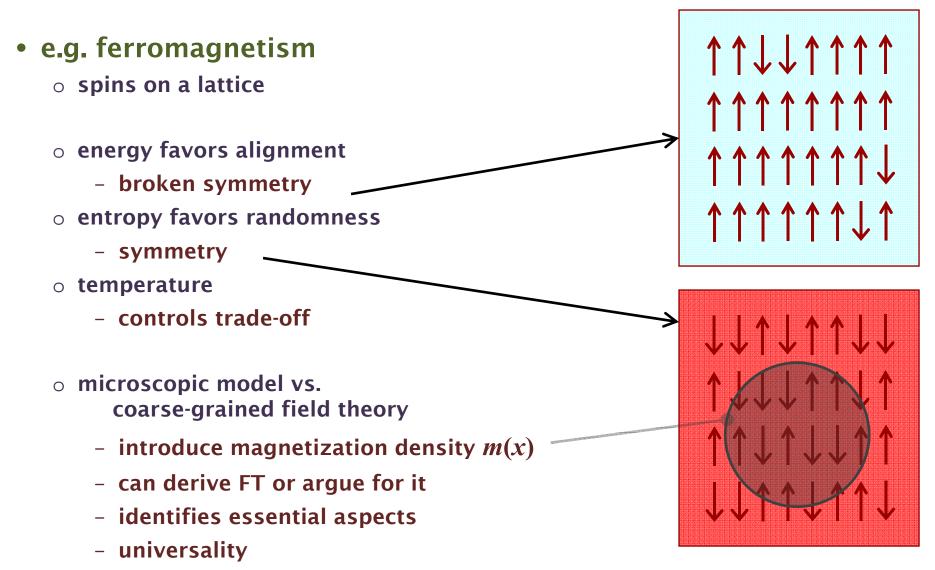


• 
$$\operatorname{try} \ \Omega ig( \mathbf{k}^0, \mathbf{k}^1, \dots, \mathbf{k}^n ig) = \left[ rac{1}{J} \sum_{j=1}^J \langle \mathrm{e}^{i \mathbf{k}^0 \cdot \mathbf{R}_j} 
angle \langle \mathrm{e}^{i \mathbf{k}^1 \cdot \mathbf{R}_j} 
angle \cdots \langle \mathrm{e}^{i \mathbf{k}^n \cdot \mathbf{R}_j} 
angle 
ight]$$

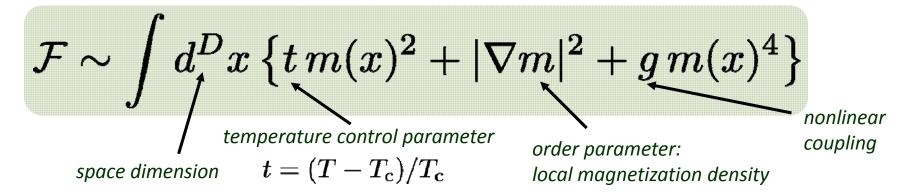
- o does discriminate, via analog of Edwards-Anderson order parameter
- o replicas yield such entities <u>naturally</u>



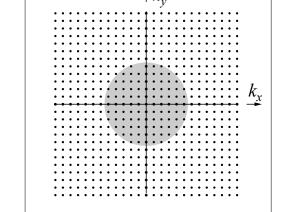
# WARM-UP: CONVENTIONAL PHASE TRANSITIONS



# FIELD THEORY APPROACH: FERROMAGNETISM



- o LW effective Hamiltonian allows for spatially varying mag den configs
- o detect FM via eq'm magn density
- T controlled breaking of spin-rotation symmetry
- o low T: eq'm m nonzero
- o high T: eq'm m zero



- instability/resolution
  - o modes in Fourier space
  - o condensation at zero k

#### Complicating factors, new & old

- fluctuations, repulsion
- crosslink constraints
- hidden spontaneous symmetry breaking

## **Edwards' strategy**

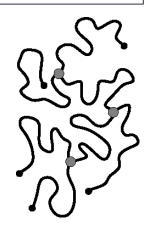
- specify constraints
- average over phase space (p.s.) of accessible polymer conformations

$$\langle \mathcal{O} \rangle = \frac{\int d(\text{p.s.}) \, e^{-\mathcal{H}(\text{p.s.})/T} \, \mathcal{O}(\text{p.s.}) \, \Delta(\text{constraints})}{\int d(\text{p.s.}) \, e^{-\mathcal{H}(\text{p.s.})/T} \, \Delta(\text{constraints})}$$

- then average <u>observables</u> over constraints
  - o replicas a smart encoding



**Sir Sam Edwards** 



## INTERMEZZO: THE REPLICA TRICK

#### Difficulty

- o shouldn't average partition function over QRVs
- o should average intensive or extensive observables
- o hard! logs or quotients
- Trick for free energy, [...] indicates quenched info average

$$[F] = -T[\ln Z] = -T\lim_{n\to 0} ([Z^n] - 1)/n$$
 uses  $z^n = e^{n \ln z} = 1 + n \ln z + \mathcal{O}(n^2)$ 

• Example: spin glasses  $\mathcal{Z}(J) = \operatorname{Tr}_S \operatorname{e}^{\sum_{j < k} (J_{jk}/T) S_j S_k}$ 

$$\begin{split} \left[ \mathcal{Z}(J)^{n} \right]_{J} &= \left[ \operatorname{Tr}_{S^{1}} \cdots \operatorname{Tr}_{S^{n}} e^{\sum_{\alpha=1}^{n} \sum_{j < k} (J_{jk}/T) S_{j}^{\alpha} S_{k}^{\alpha}} \right] \\ &= \operatorname{Tr}_{S^{1}} \cdots \operatorname{Tr}_{S^{n}} e^{\sum_{\alpha\beta} \sum_{j < k, l < m} \left[ (J_{jk}/T) (J_{lm}/T) \right] S_{j}^{\alpha} S_{k}^{\alpha} S_{l}^{\beta} S_{m}^{\beta}} \end{split}$$

#### Consequences

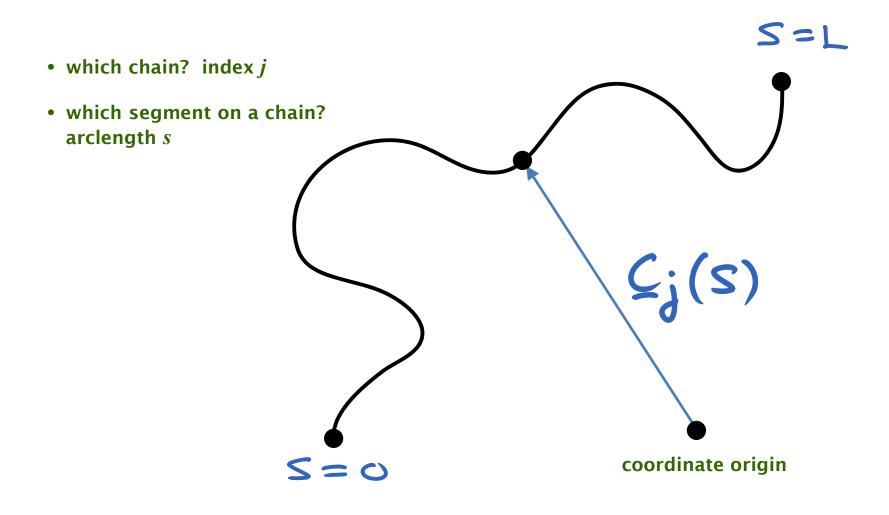
o <u>upside</u>: effective pure system; <u>downside</u>: coupled replicas

#### Physical interpretation

- $\circ$  *n* systems, same quenched disorder, independent thermal fluctuations
- o *n* measurements on same system, long time-separations

### **STRATEGY**

- o we have an order parameter
- o how can we compute it ?
  - microscopic model
    - > flexible polymers confs
    - > random constraints
  - statistical mechanics + replicas
  - derive (or argue for) a field theory
    - > a Landau-Wilson effective Hamiltonian
    - > a little strange!



$$Z \sim \int \pi \partial G \exp(-H/T) \pi \delta(G_{fm}(S_m) - G_{fm}(S_m))$$
 $Z = \int \pi \partial G \exp(-H/T) \pi \delta(G_{fm}(S_m) - G_{fm}(S_m))$ 
 $Z = \int \pi \partial G \exp(-H/T) \pi \delta(G_{fm}(S_m) - G_{fm}(S_m))$ 
 $Z = \int \pi \partial G \exp(-H/T) \pi \delta(G_{fm}(S_m) - G_{fm}(S_m))$ 

$$\int d(D(S)) P(D(S)) Z(D(S))^{n} \qquad \text{Competition}/$$

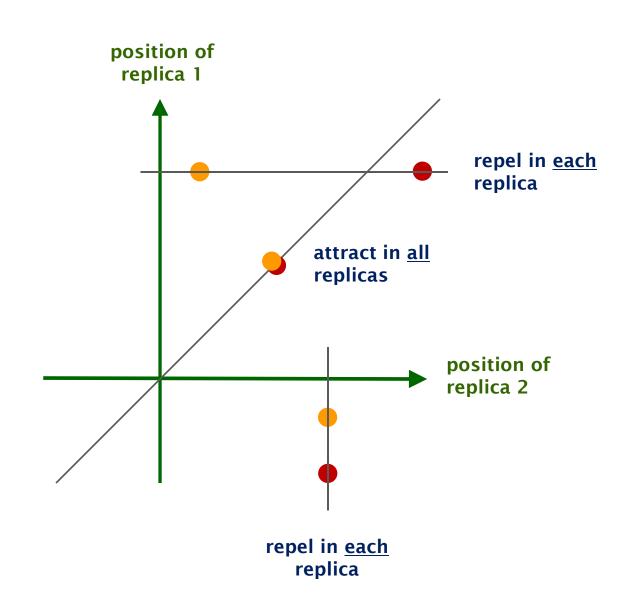
$$\int DC^{\circ}DC^{\circ}...DC^{n}$$

$$Exp - \sum_{\alpha} \sum_{j} |ds| |dc_{j}^{\alpha}/ds|^{2} \qquad \text{Particle}$$

$$x \in xp - \lambda \sum_{jk} \sum_{jk} |ds| dt \ T(c_{j}^{\alpha}(S) - c_{k}^{\alpha}(t))$$

$$x \in xp + \mu \sum_{jk} |ds| dt \ T \ T(c_{j}^{\alpha}(S) - c_{k}^{\alpha}(t))$$

$$Z \text{ Controls Coss link density}$$



# WHAT'S THE FIELD IN THE LW FT FOR RANDOM SOLIDIFICATION?

Standard technique (HS - see eg BGZ) turns semi-micro model into a FT Cronlink generated effective interaction > a real scalar field 52 but on replicated space x, x, , xn how many replicas? nearly one

# WHAT'S THE FIELD IN THE LW FT FOR RANDOM SOLIDIFICATION?

• it's precisely the "random localization detector"

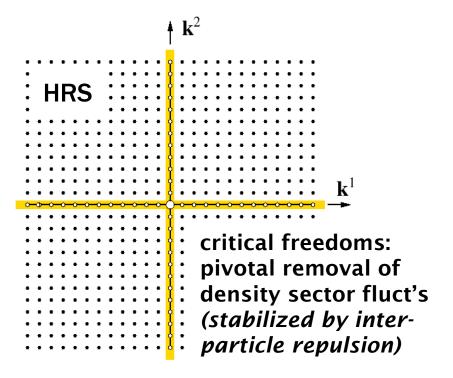
$$\Omega(\mathbf{k}^0, \mathbf{k}^1, \ldots) = \left[ J^{-1} \sum_{j=1}^{J} \langle e^{i\mathbf{k}^0 \cdot \mathbf{R}_j} \rangle \langle e^{i\mathbf{k}^1 \cdot \mathbf{R}_j} \rangle \cdots \right]$$

- or its <u>real-space</u> version  $\Omega(\mathbf{x}^0,\mathbf{x}^1,\ldots)$ 
  - o <u>oddity</u>: lives on 1+n copies of space (in the  $n \rightarrow 0$  limit) from replicas
    - body: center-of-mass of coords, macro space
    - soul: dispersion, probes localization
  - virtue: encodes rich statistical info about random solid state

#### FIELD THEORY APPROACH: RANDOM SOLIDIFICATION

$$\mathcal{F} \sim \int d^{(1+n)D}\widehat{x} \left\{ -\frac{\tau}{2}\Omega(\widehat{x})^2 + \frac{\xi_0^2}{2} |\widehat{\nabla}\Omega(\widehat{x})|^2 - \frac{g}{3!}\Omega(\widehat{x})^3 \right\}$$

- hats ?  $\hat{x} \equiv (\mathbf{x}^0, \mathbf{x}^1, \dots, \mathbf{x}^n)$
- excess constraint density: au
- object length-scale:  $\xi_0$
- cubic nonlinear coupling: g
  - derive semi-microscopically, or argue via symmetries, length-scales
  - o looks simple



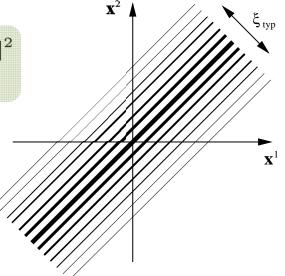
# RANDOM SOLIDIFICATION: LANDAU / MFT / CLASSICAL

$$\mathcal{F} \sim \int d^{(1+n)D}\widehat{x} \left\{ -\frac{\tau}{2}\Omega(\widehat{x})^2 + \frac{\xi_0^2}{2} |\widehat{\nabla}\Omega(\widehat{x})|^2 - \frac{g}{3!}\Omega(\widehat{x})^3 \right\}$$

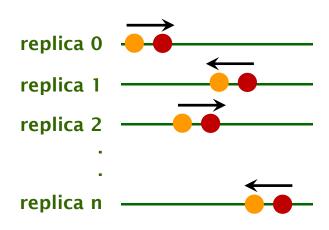
- minimize...
  - $\circ$  <u>sub</u>critical crosslink density  $\Omega(\widehat{x})=0$ 
    - all delocalized
  - o <u>super</u>critical...

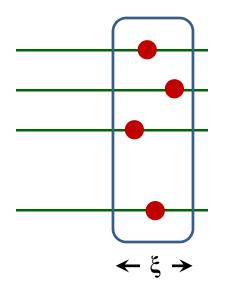
$$\Omega(\widehat{x}) \sim Q \int dz \int d\xi \, \wp(\xi) \, \mathrm{e}^{-\frac{1}{2\xi^2} \sum_{\alpha=0}^n |x^{\alpha} - z|^2}$$

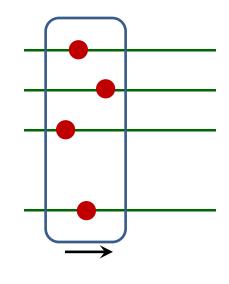
- ripple in replicated space (glide?)
- 'molecular' bound state
- random localization: peak gives *fraction*, shape gives *distribution*
- so encodes statistical information about heterogeneity of random solid state structure



#### SYMMETRIES AND SYMMETRY BREAKING







# replicas of two particle positions

effective Hamiltonian is invariant under independent translations of each replica

# pattern of symmetry breaking

replicas lock to one another - this reflects localization

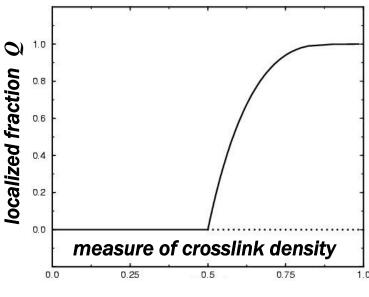
# pattern of symmetry breaking

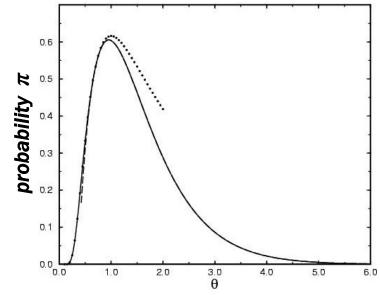
but common translation remains unbroken - this reflects macroscopic homogeneity

# LANDAU: MINIMIZING GIVES...

- $\circ$  localized fraction Q
  - recovers Erdős-Rényi
  - classical percolation exponent

- o distribution of localization lengths
  - predicts data collapse
  - universal scaling form
  - elementary derivation?

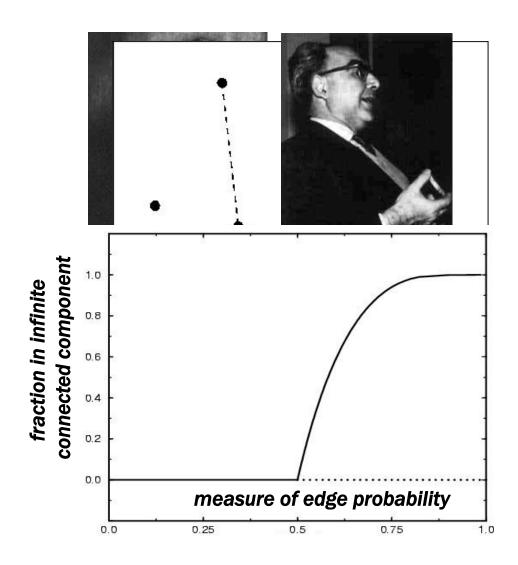




(scaled inverse square) loc. length

# MATCHES RANDOM GRAPH THEORY

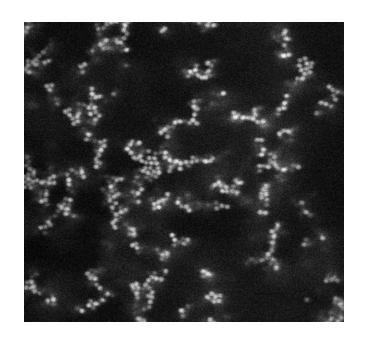
- a statistical phenomenon
- Erdős-Rényi (1960)



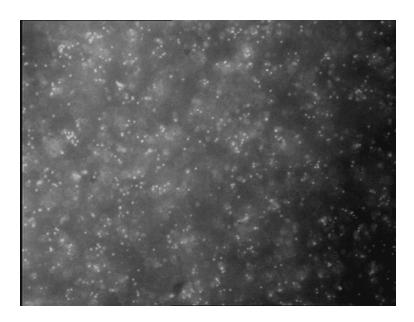
## LANDAU THEORY VS EXPERIMENTS

Dinsmore/Weitz
 (U. Mass. Amherst/Harvard)

Colloidal gels

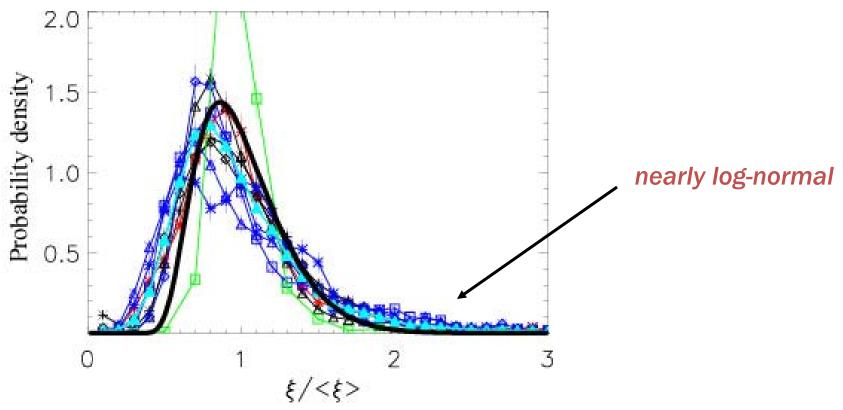


# Protein gels



- roughly μm diameter particles
- confocal microscopy
- video imaging /particle tracking
- statistical analysis of motions
- µm-scale thermal fluctuations

# LANDAU THEORY VS EXPERIMENTS



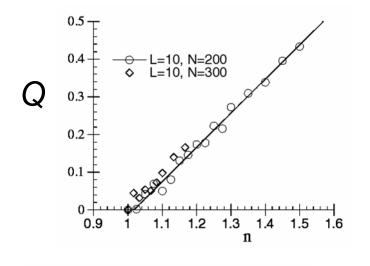
# Data on colloidal gels & protein gels (Dinsmore & Guertin, U Mass):

black: gelatin with fluorescent tracer beads
blue: particle gels by depletion attraction
red: particle gel by polycation adsorption

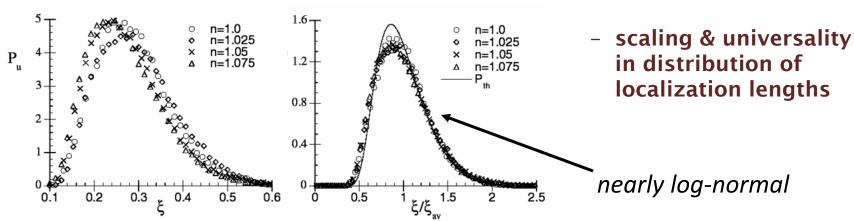
• green: colloidal crystal <a href="Theory">Theory</a>: heavy black curve

# LANDAU THEORY VS SIMULATIONS

- Barsky-Plischke MD simulations
  - o continuous transition to amorphous solid state



- N chains
- L segments
- -n crosslinks per chain
- localized fraction Q grows linearly



## MEASURING THE DETECTOR?

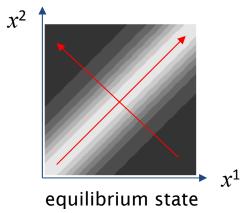
in the detector...

set most wave vectors to zero

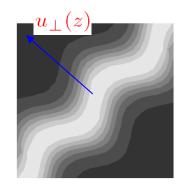
$$\Omega(\mathbf{0}, \mathbf{k}, -\mathbf{k}, \mathbf{0}, \ldots) = \left[ J^{-1} \sum_{j=1}^{J} \langle e^{i\mathbf{k} \cdot \mathbf{R}_j} \rangle \langle e^{-i\mathbf{k} \cdot \mathbf{R}_j} \rangle \right]$$

- o a part of the momentum-dependent scattering function
  - quasi-elastic ~ clustered correlator
  - incoherent ~ one particle at a time
- encodes statistical info about random solid state
  - Laplace transform of distribution of localization lengths

### **ELASTICITY OF THE RANDOM SOLID STATE**







symmetry related eq. state

Goldstone excitation

Goldstone modes, low-energy excitations

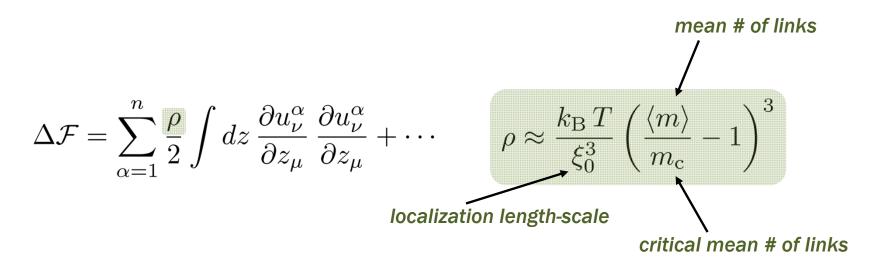
$$\Omega(\widehat{x}) \sim Q \int dz \int d\xi \, \mathcal{P}(\xi) \,\, \mathrm{e}^{-\{|x^0-z|^2 + \sum\limits_{lpha=1}^n |x^lpha - z - u^lpha(z)|^2\}/2\xi^2}$$

- o parametrized by n fields u on real space z: rippling the ripple
- o physically natural: replicas of elastic displacement fields
- o compute free energy cost
  - quadratic level gives average elastic properties
  - interactions give correlated fluctuations in elastic properties

> again: statistical information about heterogeneity of random solid state *encoded* in replica structure

# **ELASTICITY OF THE RANDOM SOLID STATE: MEAN**

 Average elastic properties obtained from quadratic-level free-energy cost of displacements



 implications of these phonon fluctuations in two dimensions & higher? (not today)

# **ELASTICITY OF THE RANDOM SOLID STATE: MEAN**

- average elastic properties obtained from quadratic-level free-energy cost of displacements
  - Lamé coefficients, stress more detailed view obtained via connection between randomly linked particle model (RLPM) & elastic phenomenology

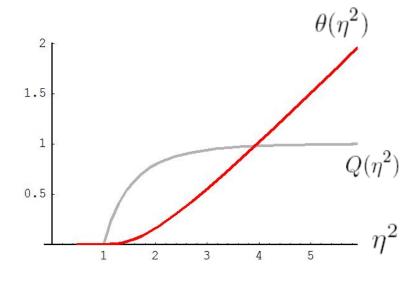
stress 
$$[\sigma_{\mu\nu}(z)] = 0$$
  
shear  $[\mu(z)] = (N/V) \theta k_{\rm B} T$   
bulk  $[\lambda(z)] = (N/V)^2 \nu^2$ 

*N/V* particle density

heta dim'less scaling function from RLPM

 $v^2$  excluded volume in RLPM

 $\eta^2 = \langle m \rangle$  mean # of links to each particle



$$\theta \equiv -\frac{1}{2} \eta^2 Q^2 + e^{-\eta^2 Q} + \eta^2 Q - 1$$

# **ELASTICITY OF THE RANDOM SOLID STATE: HETEROGENEITY**

- statistics of elastic <u>heterogeneity</u> obtained from beyond-quadratic-level displacements
  - correlations in spatial fluctuations of Lamé coefficients, get via linked particle / elastic phenomenology connection
  - key results: all correlations with stress are long-ranged, e.g.

$$[\sigma(r)\,\sigma(0)]\sim \theta T^2(N/V)/r^3$$

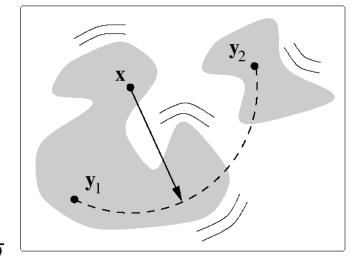
– all others are short-ranged  $\left[\mu(z)\,\mu(0)\right]_{\mathrm{con}}$ 

N/V particle density; three dimensions  $\theta$  a dim'less scaling function from RLPM

# **BEYOND LANDAU**

- fluctuations, criticality, universality
  - Ginzburg crosslink-density window

$$(\text{seg's per chain})^{-\frac{D-2}{6-D}} (\text{vol frac})^{-\frac{2}{6-D}}$$

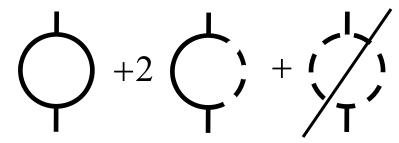


- favors short chains, dilute systems, low dimensions (cf de Gennes, 1977)
- critical dimensions: 2 & 6
- o critical state [cf Stenull-Janssen, 2001]
  - percolation questions get exactly percolation answers
  - so vulcanization field theory contains percolation
- $_{\circ}~$  field correlations  $\langle\Omega\Omega\rangle-\langle\Omega\rangle\langle\Omega\rangle$ 
  - liquid regime: mutual localization (aka clusters)
  - solid regime: not easy! correlations in motion (2D special) & structural heterogeneity, critical elasticity classes?

# PERCOLATION SECTOR

# **HRW** percolation field theory

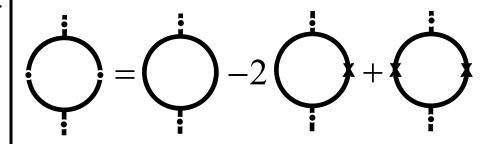
$$\int dx \left\{ \left( -\frac{\tau}{2} \phi(x)^2 + \frac{1}{2} |\nabla \phi(x)|^2 \right) - \left( -\frac{\tau}{2} \psi(x)^2 + \frac{1}{2} |\nabla \psi(x)|^2 \right) + \frac{g}{3!} \left( \phi + \psi \right)^3 \right\}$$



- ghost field sign
- $\Rightarrow$  by-hand elimination  $\Rightarrow$  -
- HRW: Houghton-Reeve-Wallace
- Peng-PMG: 1st order
- Janssen-Stenull, Peng et al: All orders

# vulcanization field theory

$$\int d\widehat{x} \left\{ -\frac{\tau}{2} \Omega(\widehat{x})^2 + \frac{1}{2} |\widehat{\nabla} \Omega(\widehat{x})|^2 - \frac{g}{3!} \Omega(\widehat{x})^3 \right\}$$



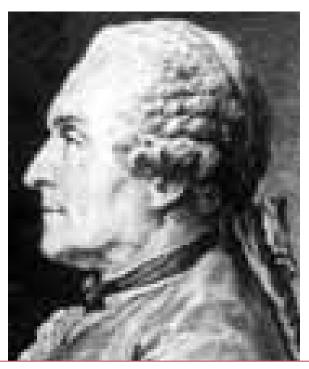
- > HRS constraint
- > momentum conservation
- > replica combinatorics
- replica limit

$$\Rightarrow$$
  $\bigcirc$ 

# Historical intermezzo (after Morawetz, 1985)

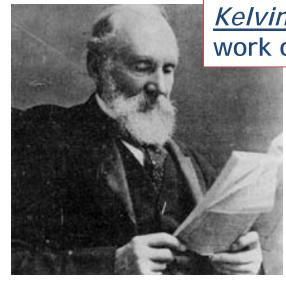


<u>Columbus</u> (Haiti, 1492): reports locals playing games with elastic resin from trees

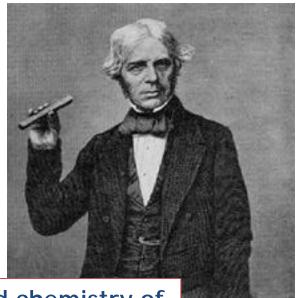


<u>de la Condamine</u> (Ecuador ~1740): latex from incisions in Hevea tree, rebounding balls; suggests waterproof fabric, shoes, bottles, cement,...

<u>Priestly</u>: erasing, coins name "rubber" (April 15, 1770)



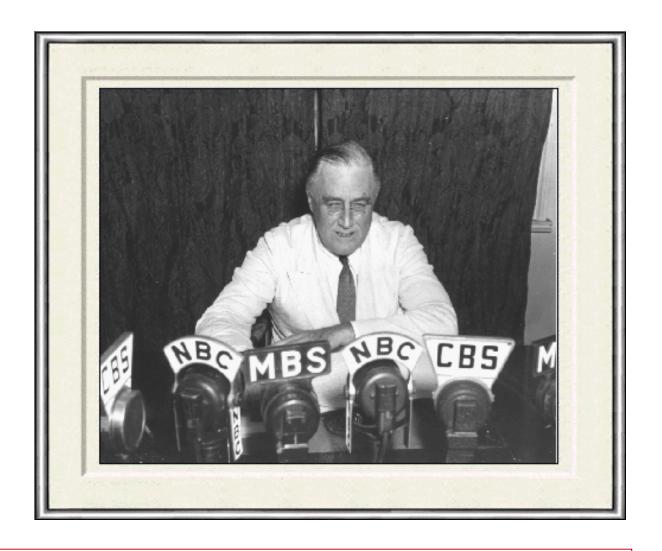
**Kelvin** (1857): theoretical work on thermal effects



<u>Faraday</u> (1826): analyzed chemistry of rubber - "...much interest attaches to this substance in consequence of its many peculiar and useful properties..."

Joule (1859): experimental work inspired by Kelvin

F. D. Roosevelt (1942, Special Committee)



"...of all critical and strategic materials... rubber presents the greatest threat to... the success of the Allied cause"

US World War II operation in synthetic rubber second in scale only to the Manhattan project





Goodyear (in Gum-Elastic and its Varieties, with a Detailed Account of its Uses, and of the Discovery of Vulcanization; New Haven, 1855):

"... there is probably no other inert substance the properties of which excite in the human mind an equal amount of curiosity, surprise and admiration. Who can reflect upon the properties of gum-elastic without adoring the wisdom of the Creator?"

<u>Dunlop</u> (1888): invents the pneumatic tyre



...which led to

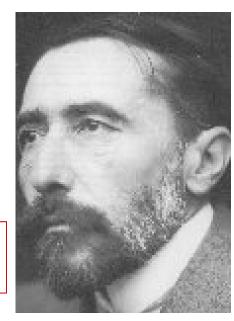
"frantic efforts to increase the supply of
natural rubber in the Belgian Congo..."

which led to

"some of the worst crimes
of man against man"

(Morawetz, 1985)

<u>Conrad</u> (1901): Heart of Darkness



# **CONCLUDING REMARKS**

- o what attracts physicists?
- o not always in obvious settings
- o random solid state
  - a standard model, its form strongly constrained
  - universality: classical & beyond
  - unified approach to...
    structure, rigidity, heterogeneity,...

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