A Toy Model for Viscosity

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Main References

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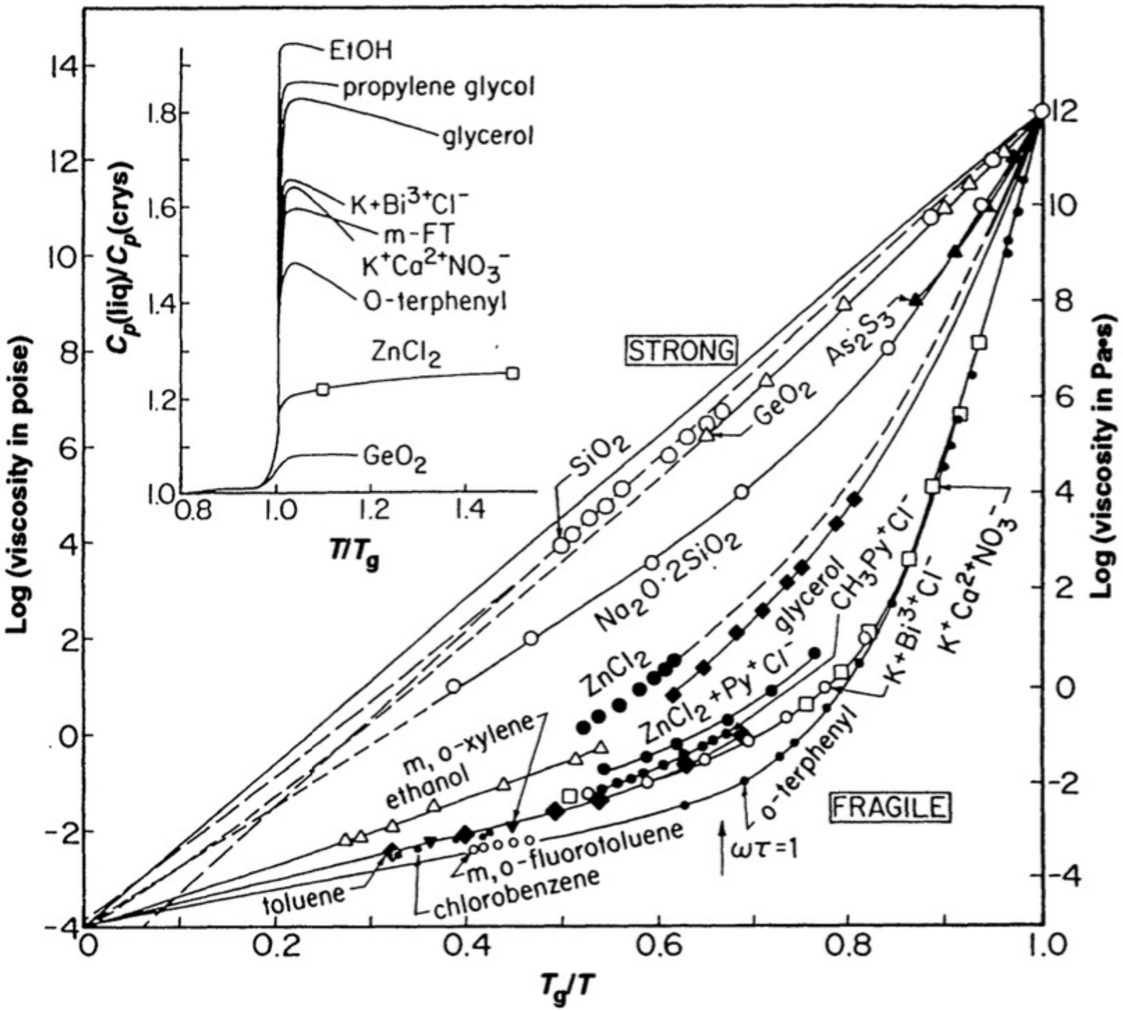
Content

Warning *This talk is reporting on a work in progress.*

1. Motivation
2. Anankeons and Phonons
3. Constructing the Model
4. Computing Maxwell's Time
5. Conclusion

I - Motivation

Motivation



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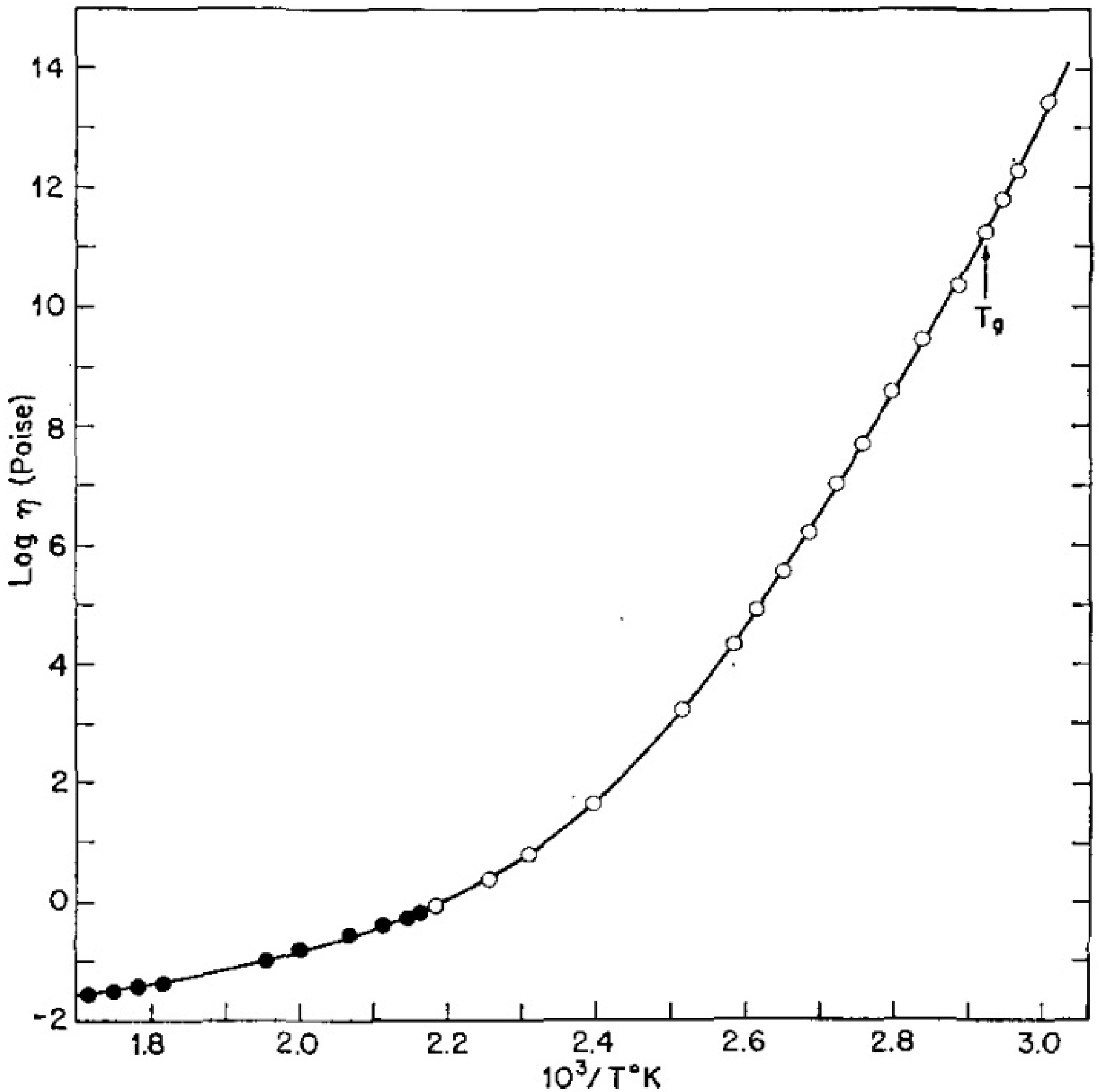
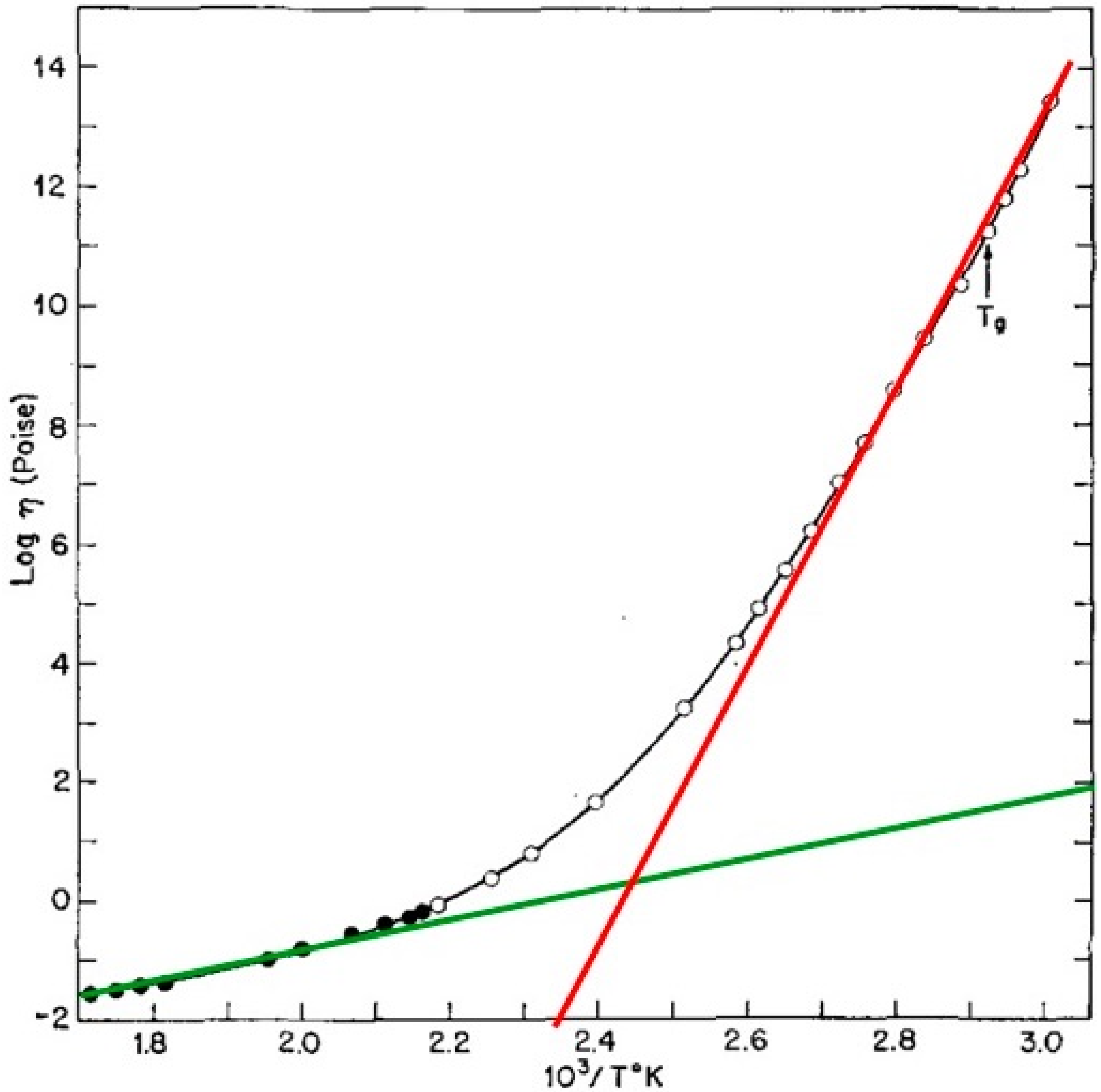
Motivation



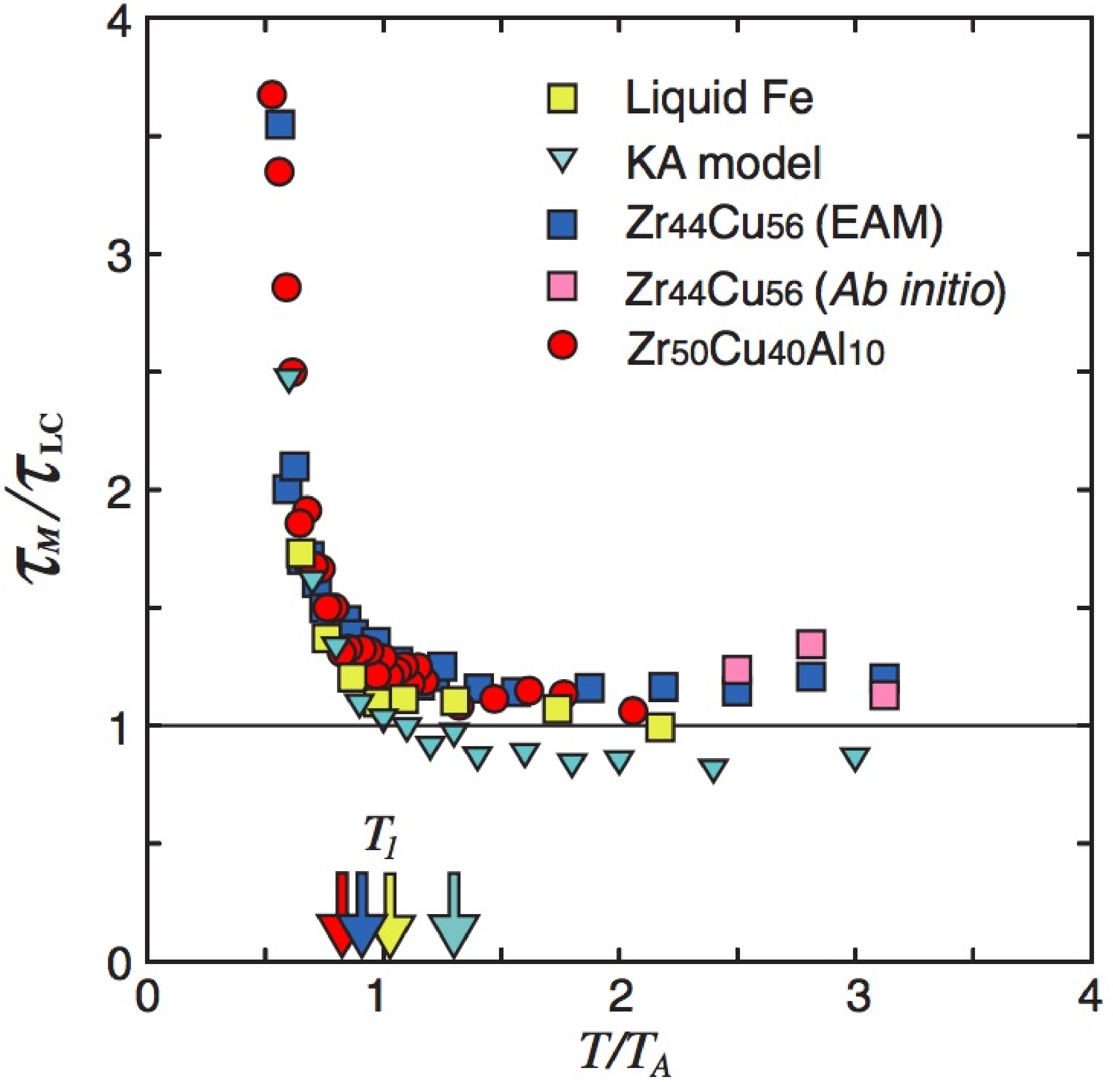
J. H. Magill, D. J. Plazek, "Physical Properties of Aromatic Hydrocarbons. II. Solidification Behavior of 1,3,5-Tri-α-Naphthylbenzene", *J. Chem. Phys.*, **45**, (1967), 3757-3769.

Motivation



Change of slope in Arrhenius behavior

Motivation



Comparison between the Maxwell relaxation time τ_M and the relaxation time τ_{LC} for local configurations.

T. Iwashita, D. M. Nicholson, T. Egami, *Phys. Rev. Lett.*, **110**, 205504-1:5, (2013).

Motivation

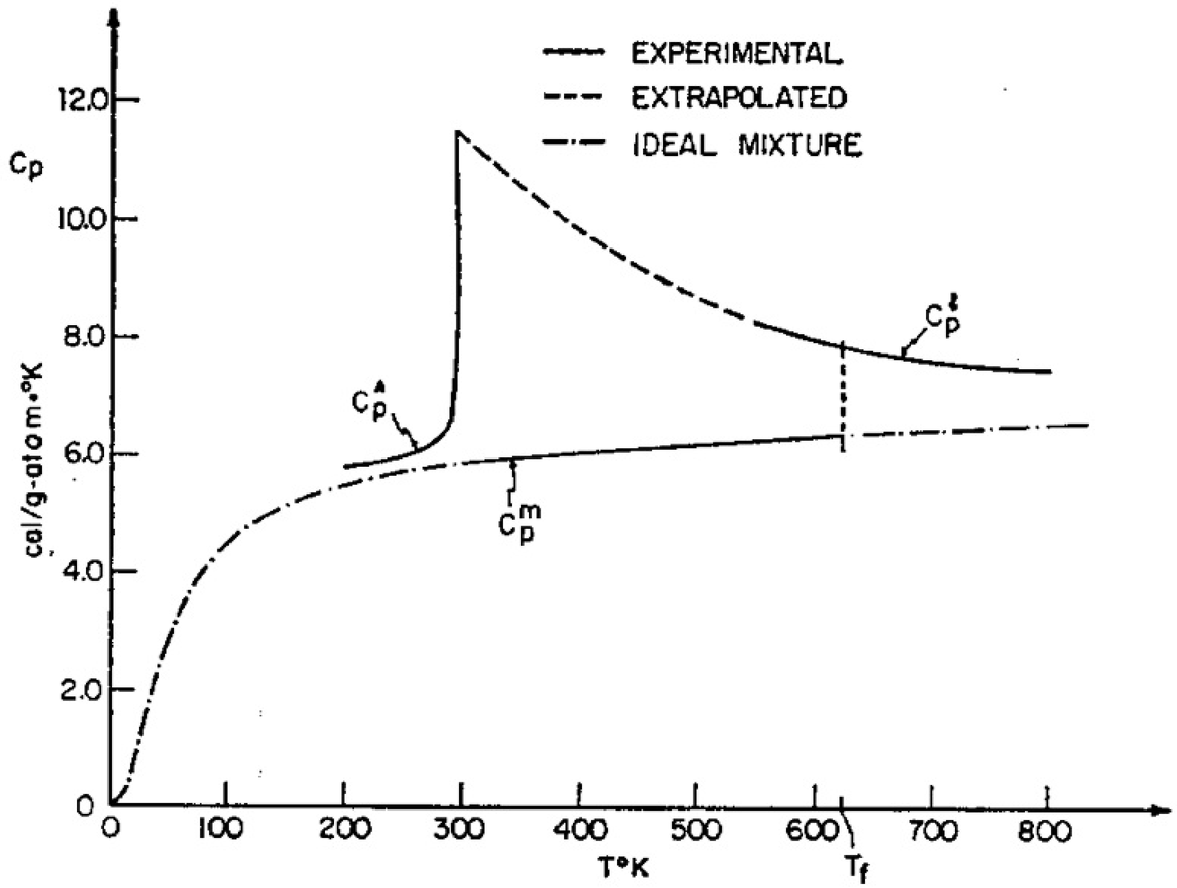


FIG. 4. Smoothed values of specific heats, C_p, of a $Au_{0.77}Ge_{0.136}Si_{0.094}$

Specific Heat for Fragile Glasses

M. H. Cohen, D. Turnbull, *J. Chem. Phys.*, **31**, 1164 (1959).

Motivation

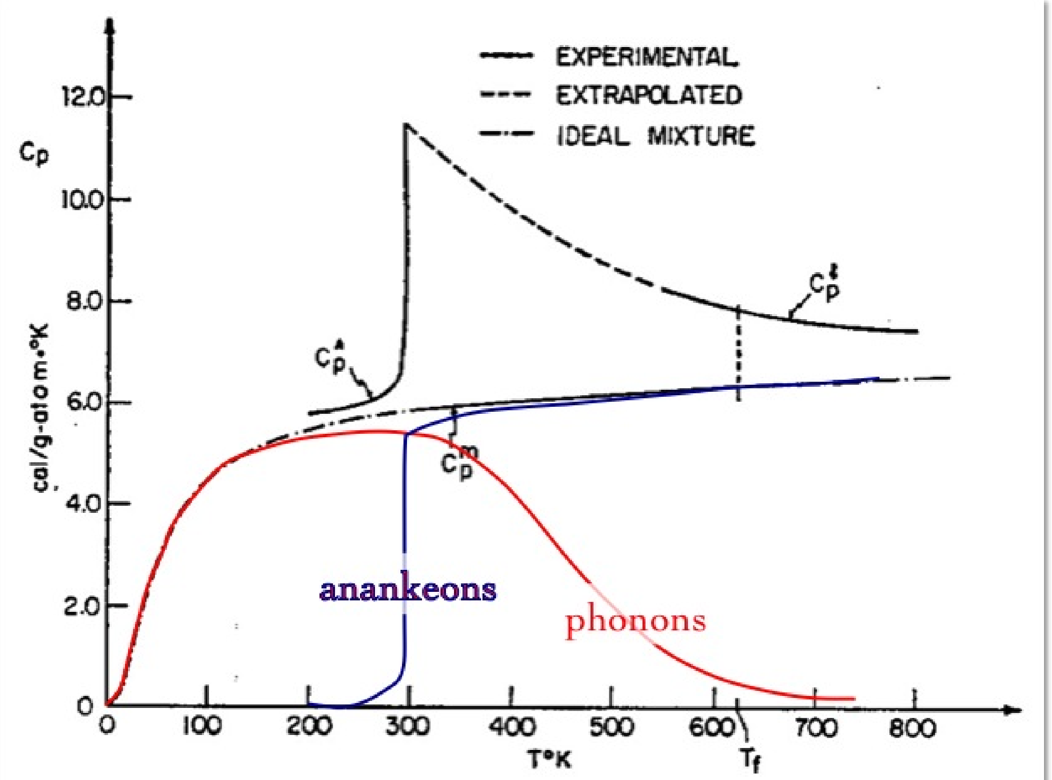


FIG. 4. Smoothed values of specific heats, C_p, of a $Au_{0.77}Ge_{0.136}Si_{0.094}$

Specific Heat for Fragile Glasses:

contributions of *phonons* & *anankeons*

Motivation

- At temperature $T > T_{co}$ larger than the *crossover* temperature, only the *anankeons* contribute to the specific heat.

- If $T_g < T < T_{co}$ phonons and anankeon *interact*.

- **Question:** *How ?*

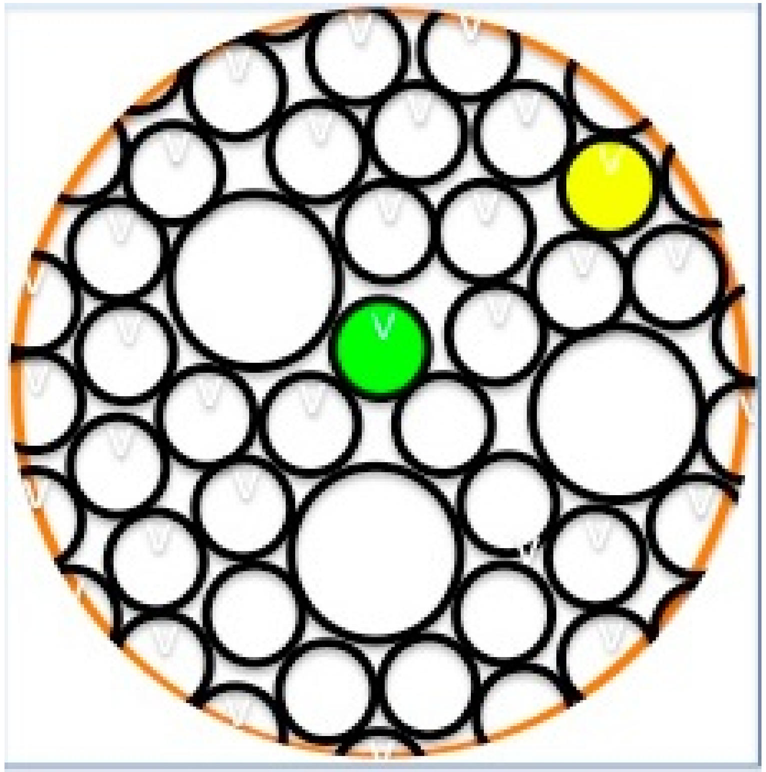
II - Anankeons and Phonons

Atomic Configurations

The set $\mathcal{L}$ of position of atomic nuclei is a *Delone set*, namely

- The *minimum distance* between atoms is $2r_0 > 0$.
- The *maximum* diameter of a *hole* without atoms is $2r_1 < \infty$.

Atomic Configurations



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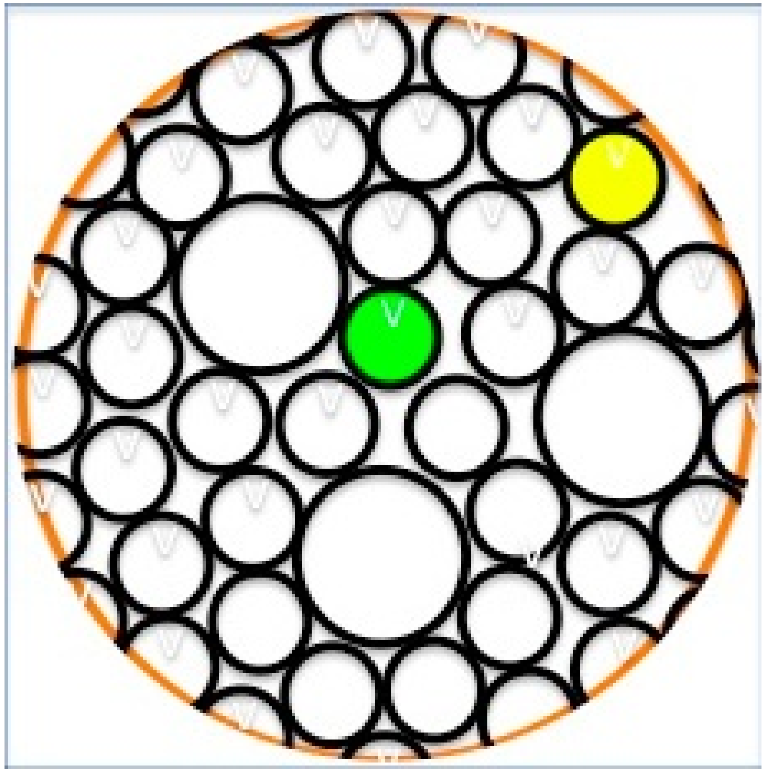

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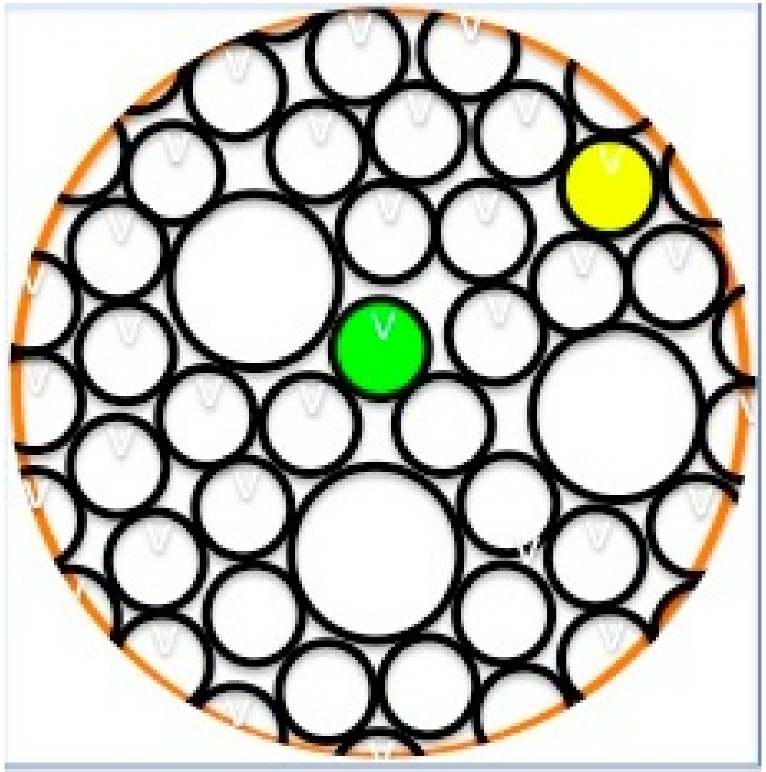

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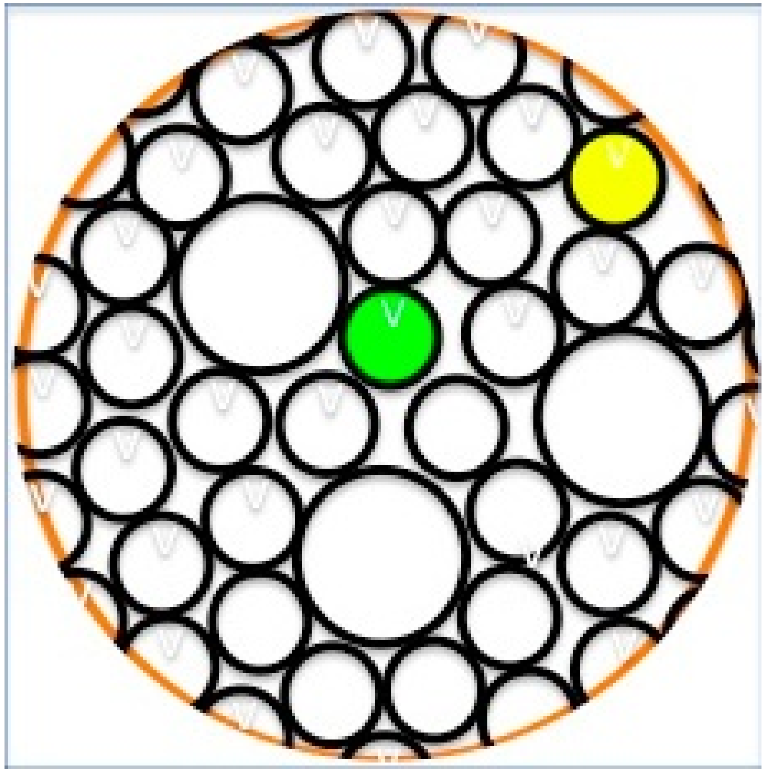

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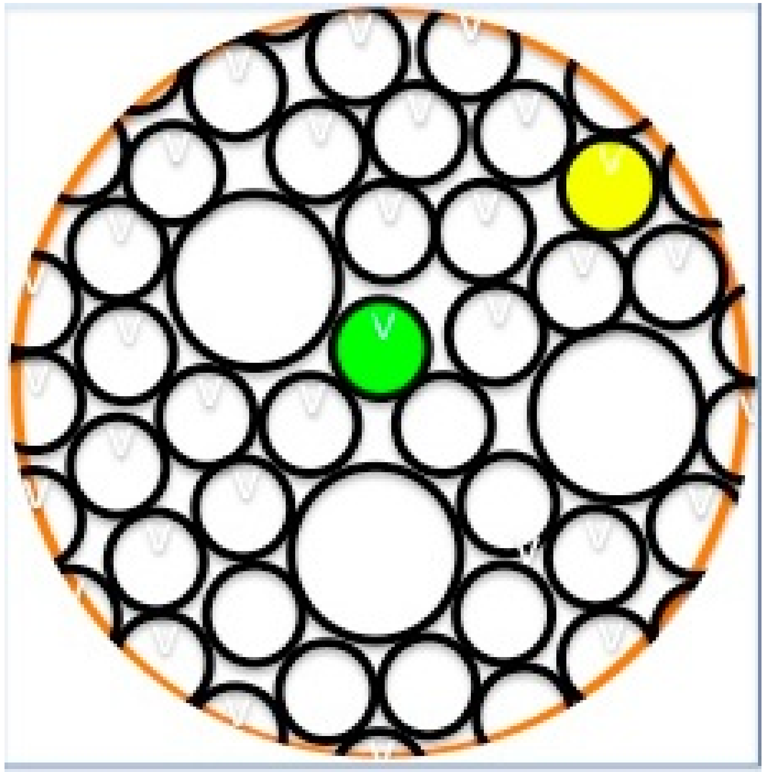

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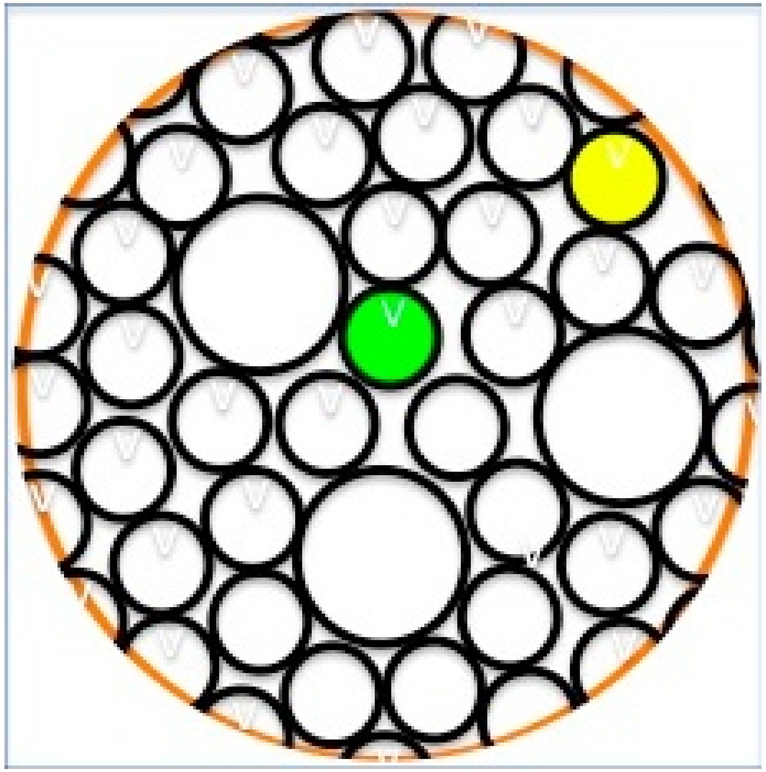

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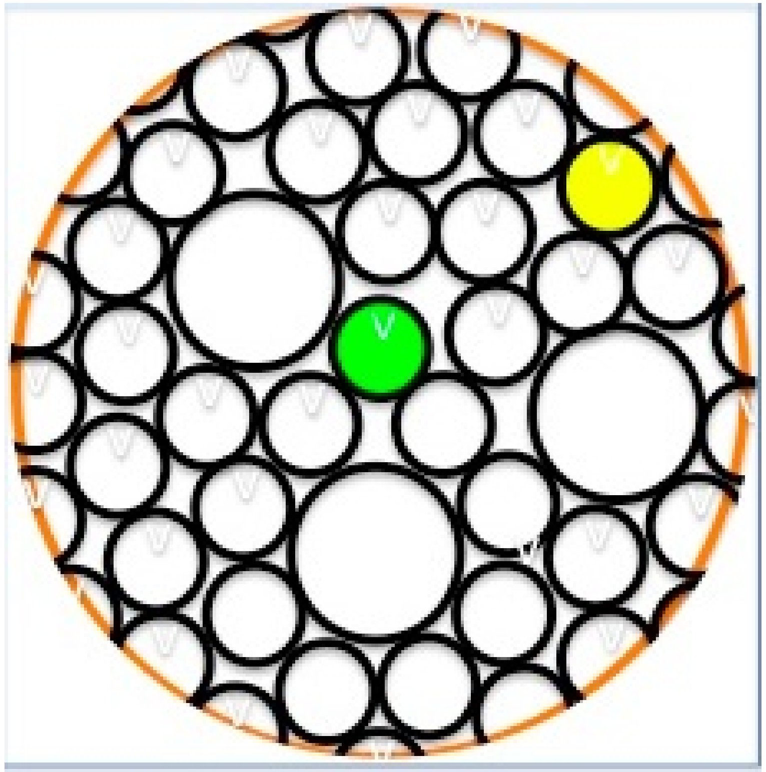

Voronoi Cells

- Let $\mathcal{L}$ be Delone. If $x \in \mathcal{L}$ its *Voronoi cell* is defined by

$$V(x) = \{y \in \mathbb{R}^d \,;\, |y - x| < |y - x'| \, \forall x' \in \mathcal{L}\,,\, x' \neq x\}$$

$V(x)$ is open. Its closure $T(x) = \overline{V(x)}$ is called the *Voronoi tile* of x

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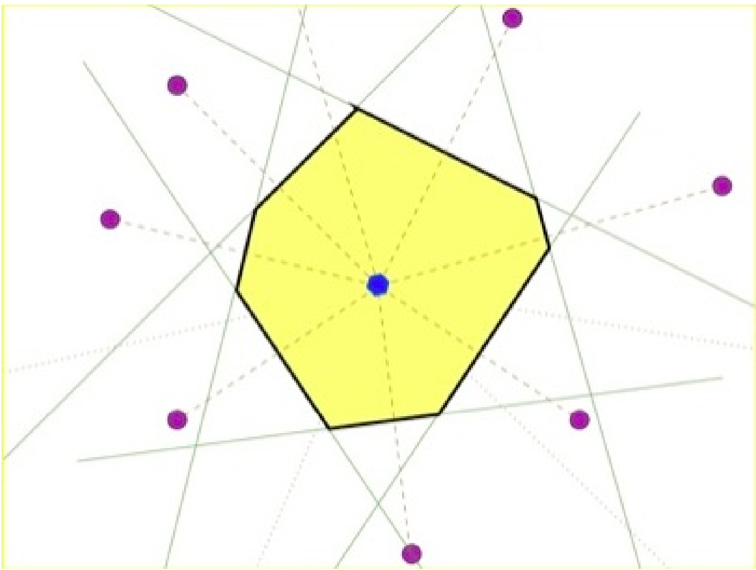

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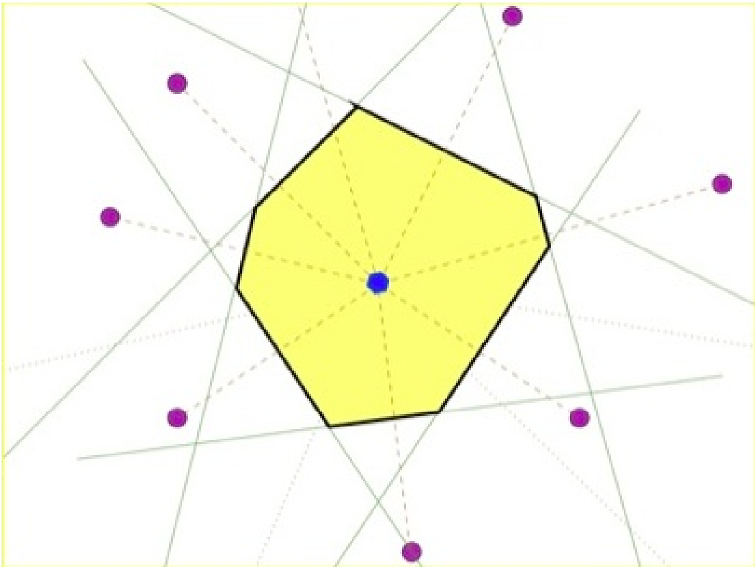

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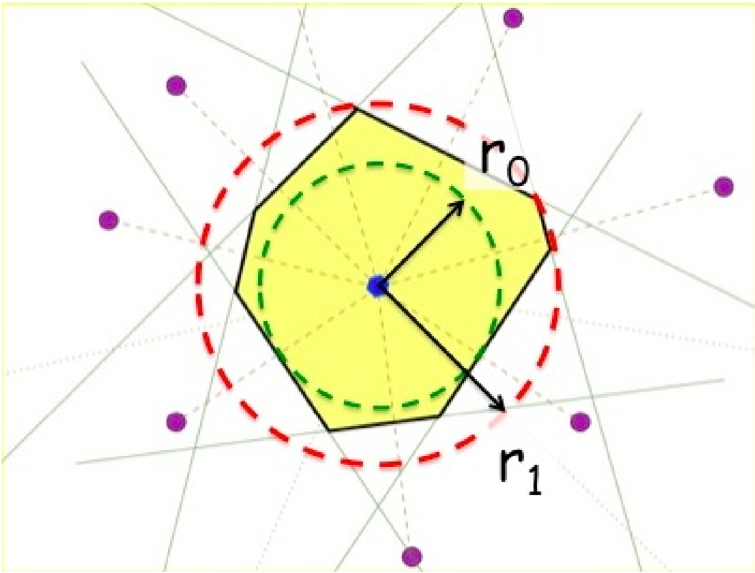
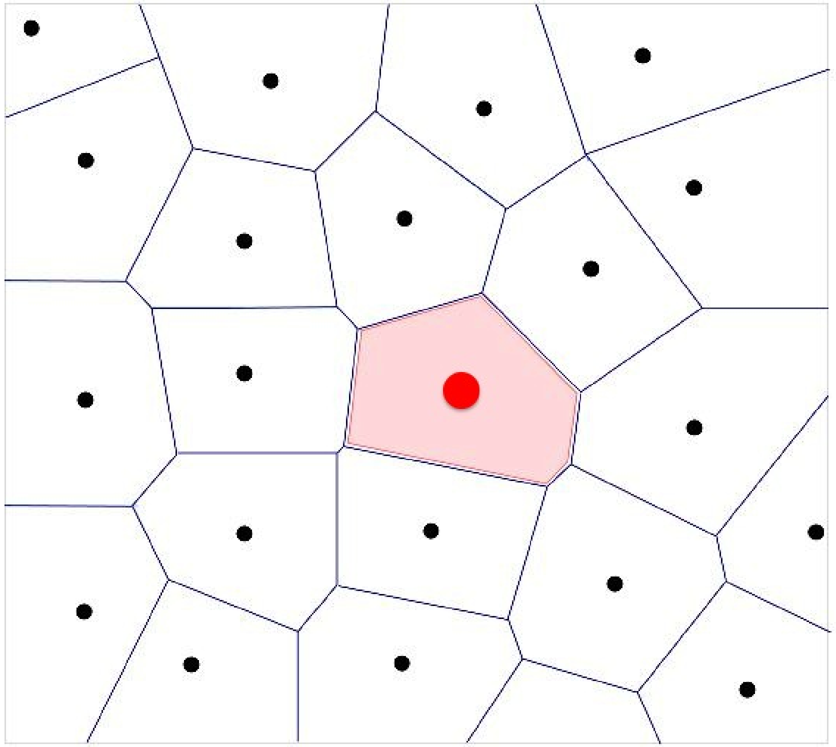


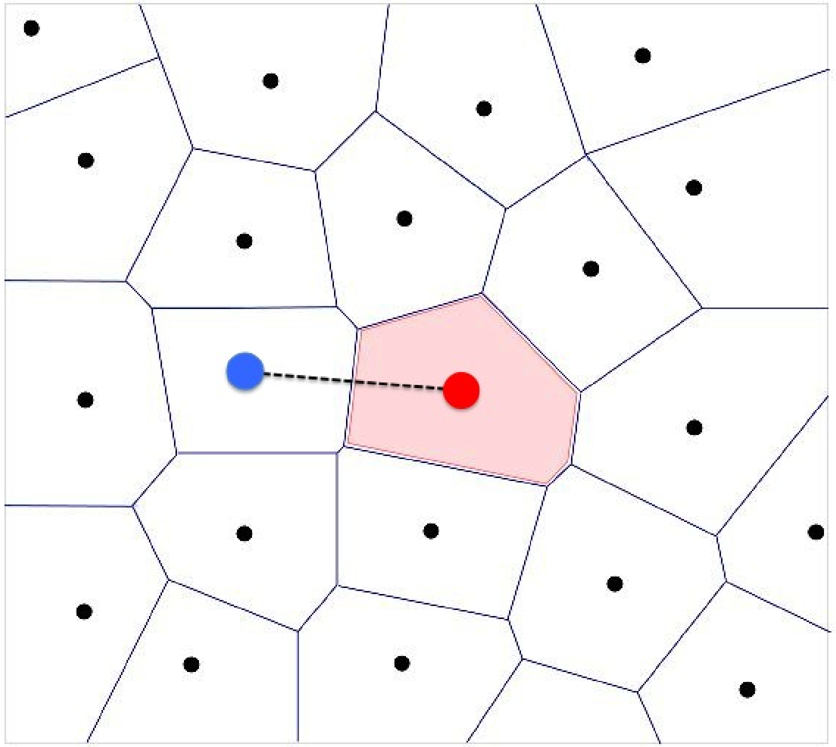
Proposition: *If $\mathcal{L}$ is Delone, the Voronoi tile of any $x \in \mathcal{L}$ is a convex polytope containing the closed ball $\overline{B(x; r_0)}$ and contained in the ball $\overline{B(x; r_1)}$*

The Delone Graph



Proposition: *the Voronoi tiles of a Delone set touch face-to-face*

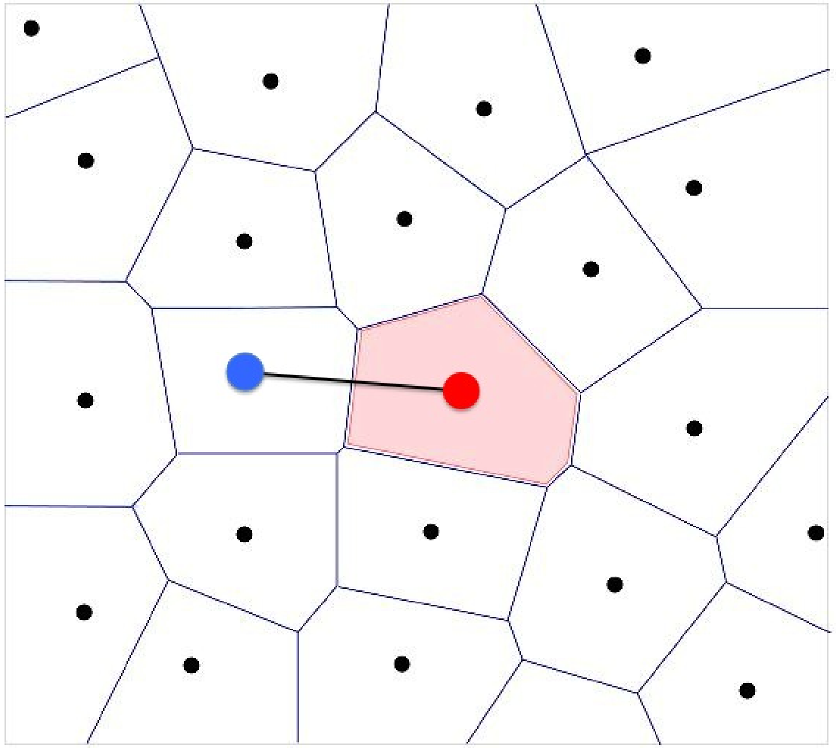
The Delone Graph



Proposition: *the Voronoi tiles of a Delone set touch face-to-face*

Two atoms are *nearest neighbors* if their Voronoi tiles touch along a face of *maximal dimension*.

The Delone Graph



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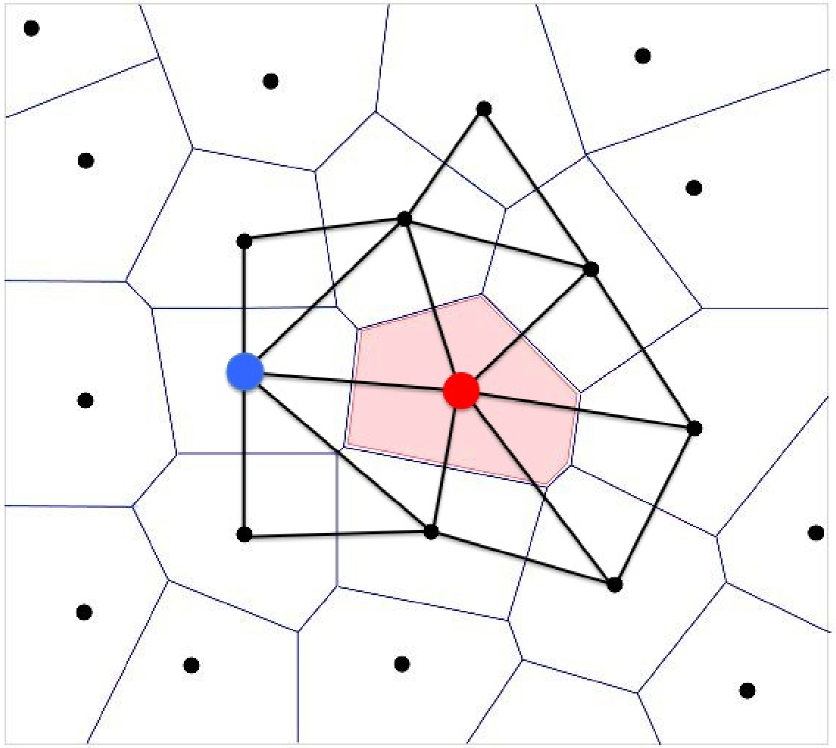
An *edge* is a pair of nearest neighbors. $\mathcal{E}$ denotes the set of edges.

The Delone Graph



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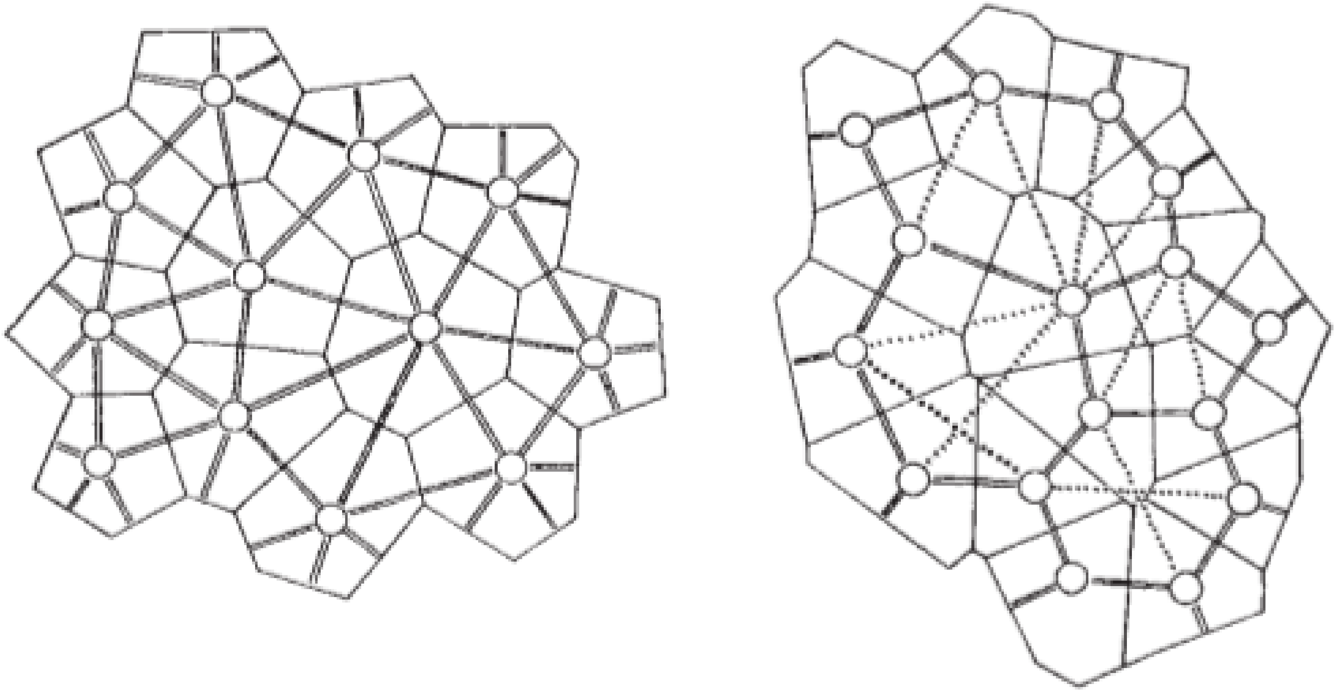
The family $\mathcal{G} = (\mathcal{L}, \mathcal{E})$ is the Delone graph.

The Delone Graph



(a) (b)

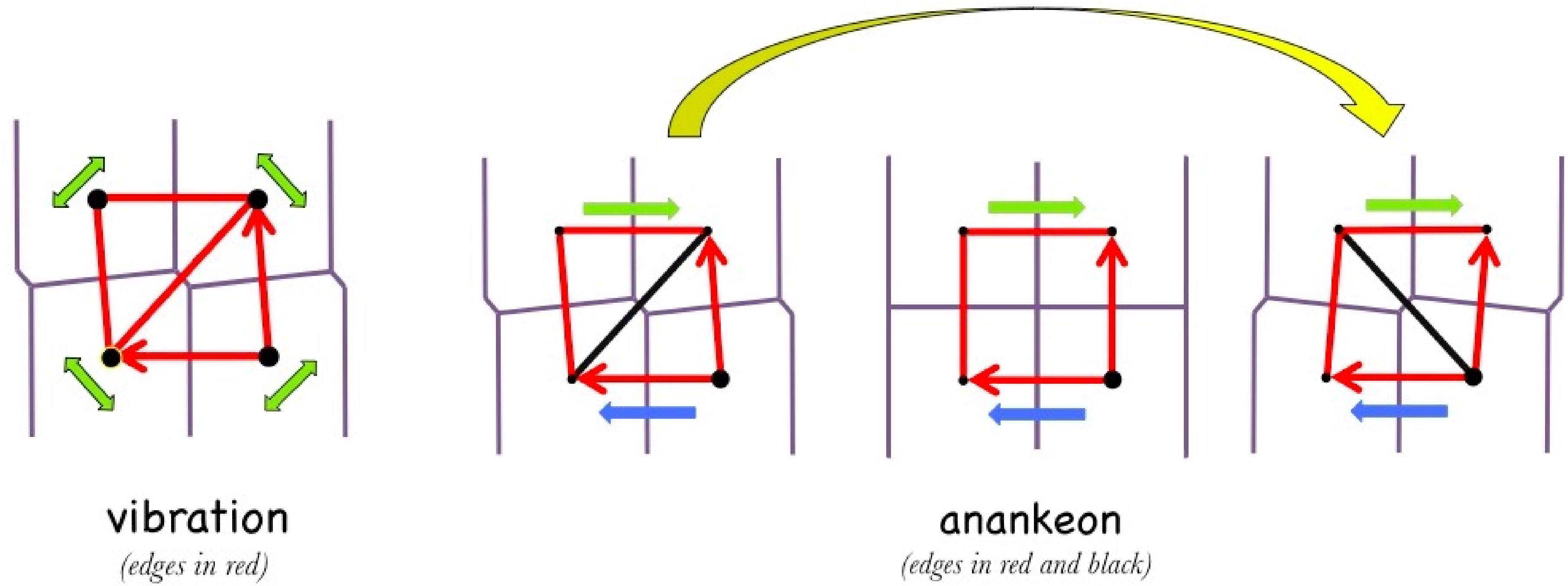
Fig. 1. Diagram of neighbourhood polyhedra, geometrical and physical, for two-dimensional arrays of points. (a) High co-ordinated; =, physical neighbours; (b) low co-ordinated; ...., geometrical neighbours

taken from J. D. Bernal, Nature, **183**, *141-147, (1959)*

The Delone Graph

Modulo graph isomorphism, the Delone graph encodes the local topology

Atomic Movements



small, deterministic,
conserves local topology

large, unpredictable, quick jump,
change the local topology

III - Constructing the Model

Harmonic Motion

- The vibration of an atom around its equilibrium position is assumed to be *harmonic*.
- To simplify further, the oscillator will be supposed to be *one-dimensional*.
- The frequency ω of the harmonic oscillator is defined by the *curvature* of the potential energy near the equilibrium position.
- Let q denotes the position of the harmonic particle *relative* to the equilibrium position. Then X will denote the *phase space vector*

$$X = \begin{bmatrix} \omega q \\ \dot{q} \end{bmatrix} = \begin{bmatrix} u \\ v \end{bmatrix}$$

Harmonic Motion

- Equation of motion

$$m\frac{d^2q}{dt^2} + kq = 0\,, \qquad \omega = \sqrt{\frac{k}{m}}\,.$$

- Equivalently

$$\frac{dX}{dt} = \omega J X\,, \qquad J = \begin{bmatrix} 0 & 1 \\ -1 & 0 \end{bmatrix}$$

- Or else

$$X(t) = e^{\omega t J} X(0) = \begin{bmatrix} \cos\omega t & \sin\omega t \\ -\sin\omega t & \cos\omega t \end{bmatrix} \begin{bmatrix} u(0) \\ v(0) \end{bmatrix}$$

Anankeon Interaction

- At *random times* $\cdots < \tau_{n-1} < \tau_n < \tau_{n+1} < \cdots$, the atom *jumps quickly* from one potential well to another one.

- Each jump will be considered as *instantaneous*.

- In the new potential well, the *curvature is different*, hence, after the time τ_n, the new frequency is ω_n.

- After the jump, the new relative *phase space position is changed* by a vector ξ_n, namely

$$X(\tau_n + 0) = X(\tau_n - 0) + \xi_n$$

Randomness: Assumptions

- The random times are *Poissonian*, namely the variables $\tau_{n+1} - \tau_n$ are *i.i.d*, with exponential distribution and average

$$\mathbb{E}\{\tau_{n+1} - \tau_n\} = \langle \tau_{n+1} - \tau_n \rangle = \tau_{LC}$$

- The frequencies ω_n are also *random* and *i.i.d*, such that

$$\mathbb{E}(\omega_n) = \omega\,, \qquad \mathbb{E}\{(\omega_n - \omega)^2\} = \sigma^2$$

- The phase-space initial positions ξ_n are also *random* and *i.i.d*, with *Gibbs distribution*

$$\mathrm{Prob}\{\xi_n \in \Lambda\} = \int_\Lambda e^{-\beta m|\xi|^2/2}\ \frac{d^2\xi}{2\pi k_B T/m}\,, \qquad \beta = \frac{1}{k_B T}$$

Correlation Function

- The goal is to compute the *stress-stress correlation function*. It is sufficient to compute

$$C_f(t) = \mathbb{E}\{f(X(t))\overline{f(X(0))}\}$$

for any complex valued function f defined on the phase space and vanishing at infinity.

- The *viscosity* is given by the Green-Kubo formula

$$\eta = \frac{V}{k_{\mathrm{B}}T}\int_0^\infty C_f(t)\,dt$$

for a suitable f

Correlation Function

- Then, the goal is to show that

$$C_f(t) \overset{t\uparrow\infty}{\sim} e^{-t/\tau_M}$$

- Hence $\eta \sim \tau_M$, which allows to interpret τ_M as the *Maxwell relaxation time*.

Correlation Function

- The *dissipative evolution* operator P_t acting on the *set of functions* f is defined by

$$P_t f(x) = \mathbb{E}\{f(X(t)) \,|X(0) = x\}$$

- Then

$$C_f(t) = \int_{\mathbb{R}^2} \overline{f(x)}\, P_t f(x)\, d^2x$$

IV - Computing Maxwell's Time

Laplace Transform

- The *Laplace transform* of $C(t)$ is defined by

$$\mathfrak{L}C(\zeta) = \int_0^\infty e^{-t\zeta}\, C(t)\, dt$$

  - The function C admits the asymptotic $C(t) \overset{t\uparrow\infty}{\sim} e^{-t/\tau_M}$

if and only if

$\mathfrak{L}C(\zeta)$ is *holomorphic* *w.r.t.* ζ in the domain $\Re\zeta > -1/\tau_M$

- In practice, $-1/\tau_M$ is the *singularity nearest to the origin* in the complex plane.

Dual Actions

- **Phase Space Rotation:** If f is a function, and $x = (u, v)$ a point in the phase space, then

$$f\left(e^{\omega t\, J} x\right) = \left(e^{-\omega t \mathbb{J}} f\right)(x), \qquad -\mathbb{J} = v\partial_u - u\partial_v$$

  Hence $\mathbb{J}$ is the *phase-space angular momentum*.

- **Phase Space Translation:** Similarly, if $\xi = (\xi^1, \xi^2)$ is a phase-space vector,

$$f(x + \xi) = \left(e^{\xi \cdot \nabla} f\right)(x), \qquad \xi \cdot \nabla = \xi^1 \partial_u + \xi^2 \partial_v$$

  Hence $\xi \cdot \nabla$ is the *phase-space momentum* along the vector ξ.

Stochastic Evolution

- Let $X(t)$ the stochastic value of the *phase-space position* at time t, with initial condition $X(0) = x$.
- If n anankeons occurred during this time then $\tau_n \leq t < \tau_{n+1}$, with $\tau_0 = 0$, so that

$$f\,(X(t)) = \left\{ \prod_{j=1}^{n} \left(e^{-(\tau_j - \tau_{j-1})\omega_{j-1}\mathbb{J}}\; e^{\xi_j \cdot \nabla} \right) e^{-(t-\tau_n)\omega_n \mathbb{J}}\; f \right\}(x)$$

- In this expression the the $\tau_j - \tau_{j-1}$'s, the ω_j's and the ξ_j's are *random, independent and identically distributed*.

Stochastic Evolution

- *Averaging* and taking the *Laplace transform* leads to

$$\mathfrak{L}P_\zeta f(x) = \int_0^\infty e^{-t\zeta}\, P_t f(x)\, dt\,, \qquad\qquad \zeta \in \mathbb{C}$$

- Equivalently

$$\mathfrak{L}P_\zeta f(x) = \mathbb{E}\left\{ \sum_{n=0}^{\infty} \int_{\tau_n}^{\tau_{n+1}} e^{-t\zeta} \prod_{j=1}^{n} \left(e^{-(\tau_j - \tau_{j-1})\omega_{j-1}\mathbb{J}}\, e^{\xi_j \cdot \nabla} \right) e^{-(t-\tau_n)\omega_n \mathbb{J}}\, f(x)\ dt \right\}$$

- **Remark:** $\tau_0 = 0$, thus $t = t - \tau_n + (\tau_n - \tau_{n-1}) + \cdots + (\tau_1 - \tau_0)$. Hence $\omega_{j-1}\mathbb{J}$ can be replaced by $\omega_{j-1}\mathbb{J} + \zeta$ and the exponential pre-factor $e^{-t\zeta}$ *disappears*.

Stochastic Evolution

- First *evaluate* the integral *over time* between τ_n and τ_{n+1}, by setting $s = t - \tau_n$

$$\int_0^{\tau_{n+1}-\tau_n} e^{-s\zeta}\, e^{-s\omega_n \mathbb{J}}\, ds = \frac{1 - e^{(\tau_{n+1}-\tau_n)(\zeta+\omega_n\mathbb{J})}}{\zeta + \omega_n \mathbb{J}}$$

- Second, *average* over the $\tau_j - \tau_{j-1}$'s, using the formula (here A is an operator)

$$\mathbb{E}_\tau \left\{ e^{-(\tau_j - \tau_{j-1})A} \right\} = \int_0^\infty e^{-s/\tau_{LC} - sA} \frac{ds}{\tau_{LC}} = \frac{1}{1 + \tau_{LC} A}$$

Stochastic Evolution

- **Reminder:** if two random variables X, Y are *stochastically independent* then $\mathbb{E}\{XY\} = \mathbb{E}\{X\}\,\mathbb{E}\{Y\}$.

- The average over the $\tau_j - \tau_{j-1}$'s gives

$$\mathfrak{L}P_\zeta f(x) = \tau_{LC} \sum_{n=0}^{\infty} \mathbb{E}\left\{ \prod_{j=1}^{n} \left(\frac{1}{1+\tau_{LC}(\zeta+\omega_{j-1}\mathbb{J})} \; e^{\xi_j\cdot\nabla} \right) \frac{1}{1+\tau_{LC}(\zeta+\omega_n\mathbb{J})} \; f(x) \right\}$$

- This gives a new averaging over a product of *independent* variables.

Stochastic Evolution

- *Averaging* over the ξ_j's can be done using (*Gibbs average is a Gaussian integral*)

$$\mathbb{E}_\xi \left\{ e^{\xi_j \cdot \nabla} \right\} = e^{k_B T \Delta / 2m}, \qquad \Delta = \nabla \cdot \nabla = \partial_u^2 + \partial_v^2$$

- *Averaging* over the ω_j's leads to defining the following operator

$$\mathbb{A}(\zeta) = \mathbb{E} \left\{ \frac{1}{1 + \tau_{LC}(\zeta + \omega_{j-1} \mathbb{J})} \right\}$$

- **Remark:** $\mathbb{A}(\zeta)$ does not depend on j since all the ω_j's have same distribution.

Stochastic Evolution

- *Inserting* into the expression of the Laplace transform leads to

$$\mathfrak{L}P_\zeta f(x) = \tau_{LC} \sum_{n=0}^{\infty} \left\{ \mathbb{A}(\zeta)\, e^{k_{\mathrm{B}} T \Delta / 2m} \right\}^n \mathbb{A}(\zeta)\, f(x)$$

$$= \tau_{LC} \frac{1}{1 - \mathbb{A}(\zeta)\, e^{k_{\mathrm{B}} T \Delta / 2m}} \mathbb{A}(\zeta)\, f(x)$$

- **Questions:**
  - How do we evaluate this function ?
  - How can we compute the *domain of analyticity* in ζ ?

Angular Momentum

- **Trick:** the operator $\mathbb{A}(\zeta)$ is a function of the phase-space *angular momentum* $\mathbb{J}$.
- The *polar coordinates* in the $2D$-phase space are given by

$$\begin{cases} u = r\cos\theta \\ v = r\sin\theta \end{cases} \qquad \Leftrightarrow \qquad \begin{cases} r^2 = u^2 + v^2 \\ \tan\theta = v/u \end{cases}$$

- It follows that

$$\mathbb{J} = \frac{\partial}{\partial\theta}$$

Angular Momentum

- Consequently the *eigenvalues* of $\mathbb{J}$ are given by $\imath\ell$ with $\ell = 0, \pm 1, \pm 2, \cdots$, namely $\ell \in \mathbb{Z}$.
- The eigenfunctions have the form

$$g_\ell(r,\theta) = \hat{g}_\ell(r)\, e^{\imath\ell\theta}$$

- *Projecting* a function f onto the eigenspace of eigenvalue $\imath\ell$ is given by

$$\Pi_\ell f(r,\theta) = e^{\imath\ell\theta} \int_0^{2\pi} f(r,\theta)\, e^{-\imath\ell\theta}\, \frac{d\theta}{2\pi}$$

- Hence the *spectral decomposition* gives

$$\mathbb{J} = \sum_{\ell\in\mathbb{Z}} \imath\, \ell\, \Pi_\ell$$

Angular Momentum

- Any function of $\mathbb{J}$ can be written as

$$F(\mathbb{J}) = \sum_{\ell \in \mathbb{Z}} F(\imath\ell)\, \Pi_\ell$$

- It leads to

$$\mathbb{A}(\zeta) = \sum_{\ell \in \mathbb{Z}} a_\ell(\zeta)\, \Pi_\ell$$

with $a_\ell(\zeta)$ a complex number given by

$$a_\ell(\zeta) = \mathbb{E}_\omega \left\{ \frac{1}{1 + \tau_{LC}(\zeta + \imath\ell\omega_j)} \right\}$$

Angular Momentum

- **New Trick:** the operator Δ commutes with $\mathbb{J}$, more precisely, the polar decomposition gives

$$\Delta = -p_r^2 + \frac{\mathbb{J}^2}{r^2} \qquad\qquad -p_r^2 = \partial_r^2 + \frac{1}{r}\,\partial_r$$

- This gives

$$\mathfrak{L}P_\zeta = \tau_{LC} \sum_{\ell \in \mathbb{Z}} \frac{1}{1 - a_\ell(\zeta) e^{-k_B T (p_r^2 + \ell^2/r^2)}}\, a_\ell(\zeta)\, \Pi_\ell$$

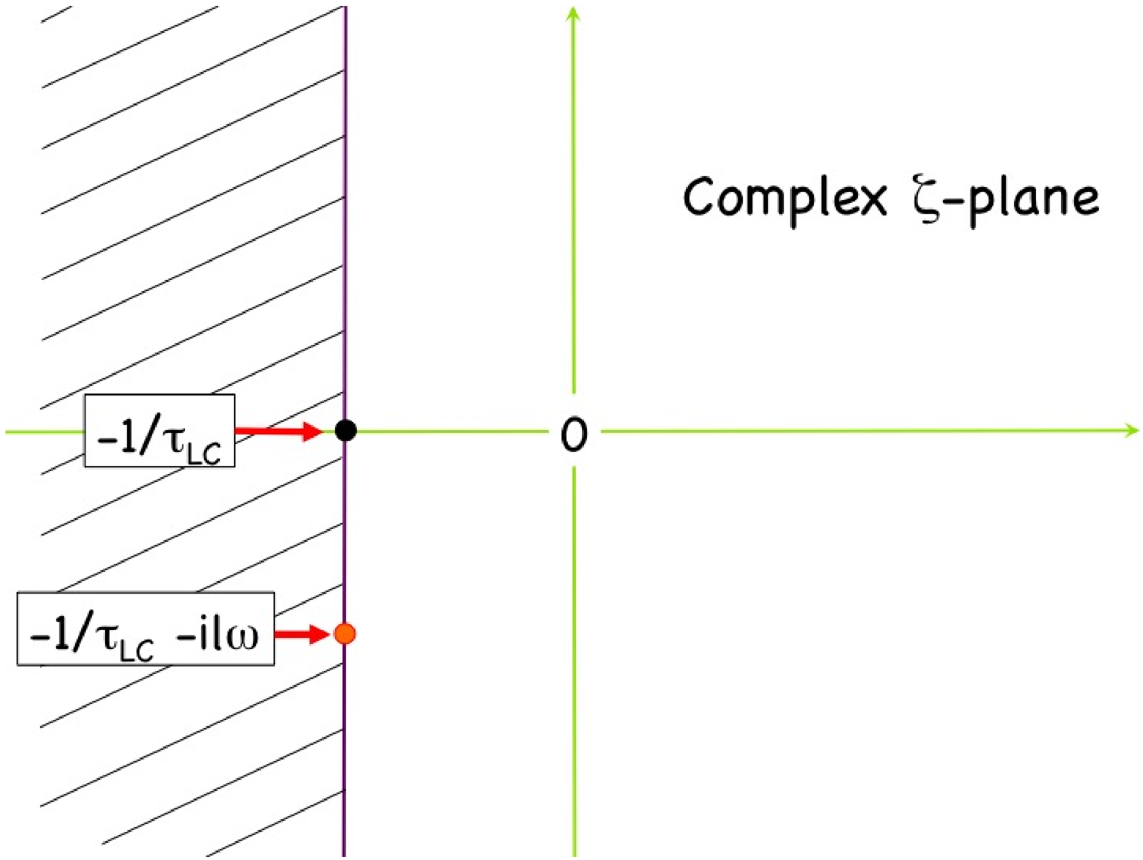
Analyticity

- Remark that if $\omega > 0$ the function $(1 + \tau_{LC}(\zeta + \imath\ell\omega))^{-1}$ admits a pole at

$$\zeta = -\frac{1}{\tau_{LC}} - \imath\ell\omega$$

- It follows that $a_\ell(\zeta)$ is analytic in $\Re(\zeta) > -1/\tau_{LC}$
- The operator $p_r^2 + \ell^2/r^2$ is positive, and its spectrum is the *entire positive real line*. Hence, $\mathfrak{L}P_\zeta$ is analytic in the domain for which there is no $\ell \in \mathbb{Z}$ nor any $p \geq 0$ such that $a_\ell(\zeta) = e^{p^2} \geq 1$.
  It means the set of ζ must satisfy $a_\ell(\zeta) \notin [1, \infty)$

Analyticity



The *domain of analyticity* of $a_l(\zeta)$ is given by $\Re\zeta > -1/\tau_{LC}$

Analyticity

- The *correlation function* for $X(t)$ involves only the angular momentum $\ell = \pm 1$.
- Assume that the distribution of ω_j is *uniform* with average ω and variance σ. Then the ω_j's are uniformly distributed in the interval $[\omega - \sqrt{3}\,\sigma, \omega + \sqrt{3}\,\sigma]$ and

$$a_{\pm 1} = \frac{1}{2\imath\sqrt{3}\tau_{LC}\sigma}\,\ln\left(\frac{1+\tau_{LC}(\zeta+\imath\omega+\imath\sqrt{3}\sigma)}{1+\tau_{LC}(\zeta+\imath\omega-\imath\sqrt{3}\sigma)}\right)$$

- If $\zeta + \imath\omega$ is *not real*, then the imaginary part of the *r.h.s.* is non zero, so that $a_{\pm 1} \notin [1, \infty)$.

Analyticity

- If $\xi = \zeta + \imath\omega$ is *real* then

$$a_{\pm 1} = \frac{1}{2\imath\sqrt{3}\tau_{LC}\sigma}\ \ln\left(\frac{1+\tau_{LC}(\zeta+\imath\omega+\imath\sqrt{3}\sigma)}{1+\tau_{LC}(\zeta+\imath\omega-\imath\sqrt{3}\sigma)}\right) = \frac{\theta}{\sqrt{3}\tau_{LC}\sigma}$$

where

$$\tan\theta = \frac{\sqrt{3}\tau_{LC}\sigma}{1+\tau_{LC}\xi} \qquad |\theta| < \frac{\pi}{2}$$

Maxwell Relaxation Time

- After some algebra, this gives

$$\tau_M = \begin{cases} \tau_{LC} & \text{if} \quad \sqrt{3}\tau_{LC}\sigma \geq \pi/2 \\ \tau_{LC}\left(1 - \dfrac{\sqrt{3}\tau_{LC}\sigma}{\tan\sqrt{3}\tau_{LC}\sigma}\right)^{-1} > \tau_{LC} & \text{otherwise} \end{cases}$$

- Using *a uniform distribution* of oscillator frequencies is a reasonable approximation. If $\tau_{LC}\,\sigma$ decreases to zero as $T \downarrow 0$, then
  - This gives a *crossover* temperature T_{co} above which the *anankeon dominates* and $\tau_{LC} = \tau_M$ for $T \geq T_{co}$
  - If $T < T_{co}$, the *phonons resist* and $\tau_M/\tau_{LC} > 1$.

Maxwell Relaxation Time

- The *variance* σ of the random frequencies depend upon the *modification of the local landscape* by anankeons. However, each anankeon involves several atoms, at least $d+2$ atoms in dimension d. So the landscape is modified in a region that might be large compared with the mean atomic distance.
- For this reason, σ is expected to be proportional to a Gibbs factor, namely to follow also an *Arrhenius law*

$$\sigma = \sigma_\infty \, e^{-W_v/k_B T} \qquad \sigma_\infty = \lim_{T \uparrow \infty} \sigma(T)$$

- **Question:** *What is the meaning of* W_v *?*

Maxwell Relaxation Time

- Similarly the local configuration time τ_{LC} is also given by a similar expression, thanks to *Kramers formula*, where now W is the *potential energy barrier* to be crossed when an anankeon occurs

$$\tau_{LC} = \tau_\infty \; e^{W/k_B T} \qquad\qquad \tau_\infty = \lim_{T\uparrow\infty} \tau(T)$$

- If the hypothesis made on σ is correct, then

$$K(T) = \tau_{LC}(T)\sigma(T) = K_\infty \, e^{-(W_v - W)/k_B T} \qquad\qquad K_\infty = \sigma_\infty \, \tau_\infty$$

The condition $K(T) \stackrel{T\downarrow 0}{\rightarrow} 0$ *requires* $W_v > W$

Maxwell Relaxation Time

- Hence, if $T_g < T < T_{co}$

$$\frac{\tau_M}{\tau_{LC}} = \frac{1}{1 - \frac{\sqrt{3}K(T)}{\tan(\sqrt{3}K(T))}} \underset{\sim}{T\downarrow 0} \frac{e^{2(W_v - W)/k_B T}}{K_\infty^2}$$

- Similarly the *crossover temperature* is reached whenever $\sqrt{3}\tau_{LC}\sigma = \pi/2$, which gives

$$(W_v - W) = \frac{k_B T_{co}}{10.2}$$

V - Conclusion

To Summarize

- The liquid phase of *fragile glasses* is dominated by *anankeons* at least above the crossover temperature $T > T_{co}$.
- If $T_g < T < T_{co}$, the *phonon-anankeon interaction* becomes essential to explain the difference between the Maxwell and the local configuration times. Hence the change of the Arrhenius behavior of the viscosity is explained through a *dynamical effect*. As the temperature decreases, phonons become more coherent and *limit the dissipative* effect of the anankeons.
- The crossover temperature is related to the difference $W_s - W$ between the activation energies associated with the *phonon frequency fluctuation* and the *anankeon potential barrier*.