We have generated sets of (x,y,z) positions for various times at various given thermodynamic conditions (N,V,T,P). Today we focus mostly on time dependence !

Correlation functions and linear response theory

- Mean square displacement and diffusion
- Van Hove correlation functions
- Intermediate scattering functions
- Linear response theory, time correlations
- Applications

A) MEAN SQUARE DISPLACEMENT AND DIFFUSION

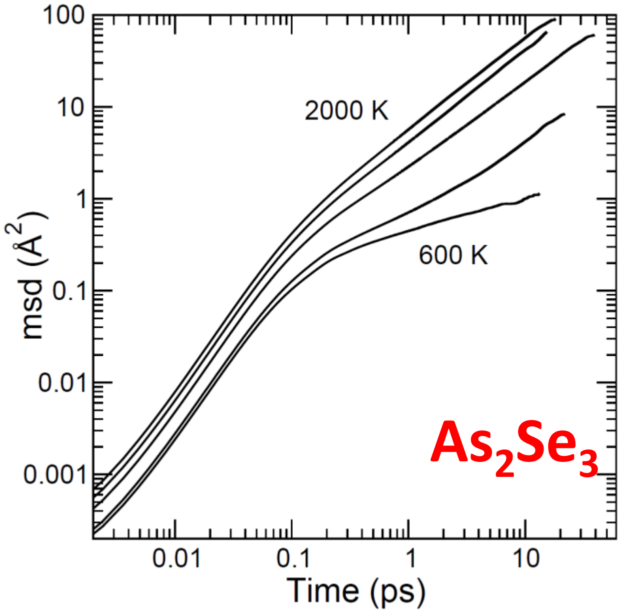
The mean square displacement is defined as

$$\langle r^2(t)\rangle = \frac{1}{N_\alpha}\sum_{i=1}^{N_\alpha}\langle|r_i(t)-r_i(0)|^2\rangle,$$

- performed in NVE or NVT.

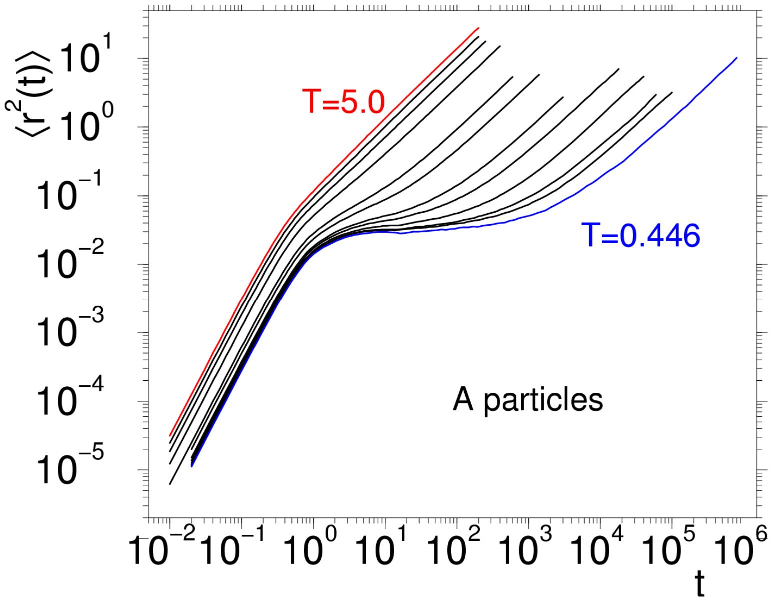
- do not use periodic boundary conditions

Gives a direct description of the dynamics.



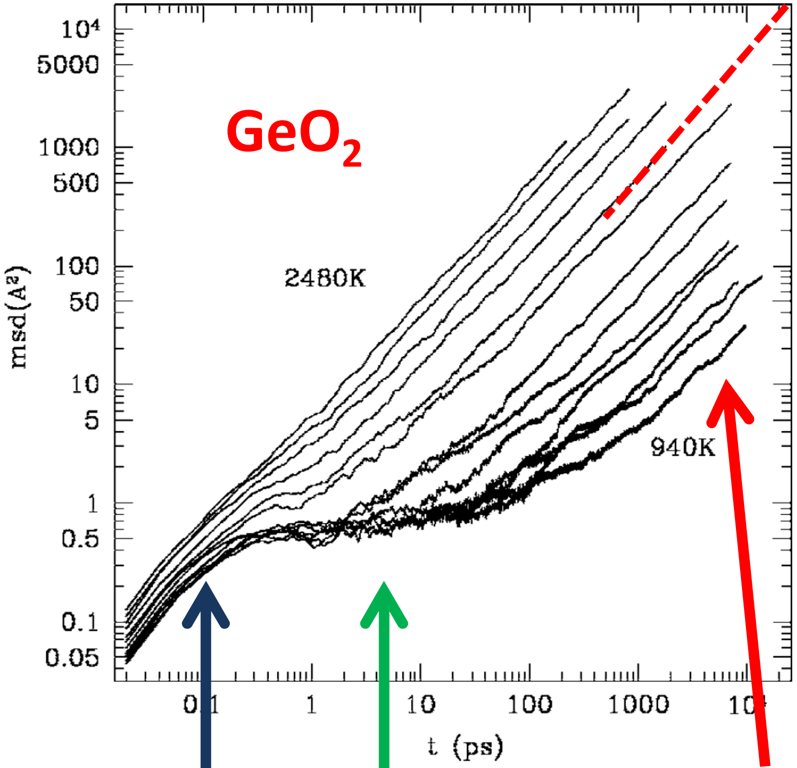
Bauchy et al., PRL 2013

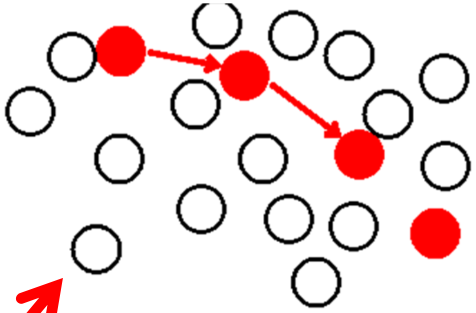
A-B Lennard-Jones liquid

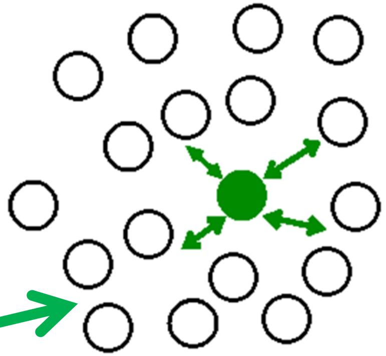


Kob PRE 2000

More insight into the msd…







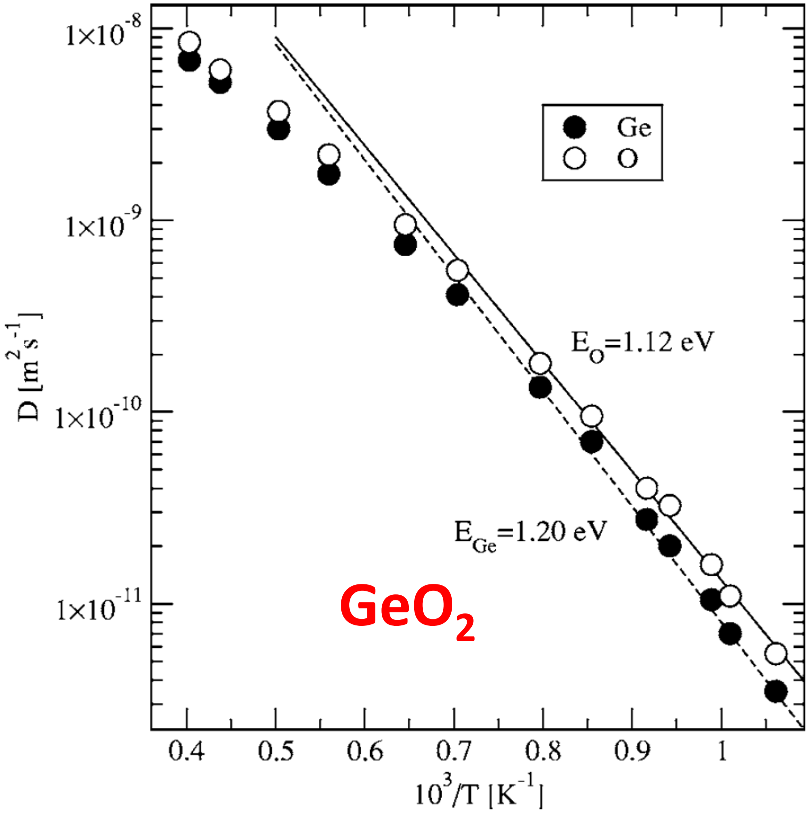
Diffusive régime
Long times

Ballistic régime
msd~t^2
Short time

Caging régime (LT)

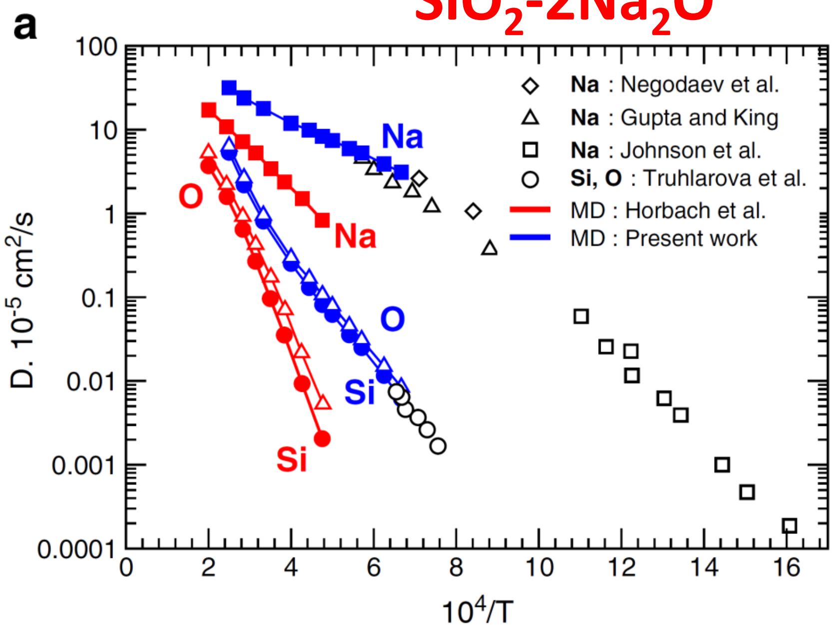
Behavior with temperature

Usually, a gentle Arrhenius behavior : $D=\exp[-E_A/k_BT]$



Micoulaut et al. PRE 2006

SiO_2-$2Na_2O$

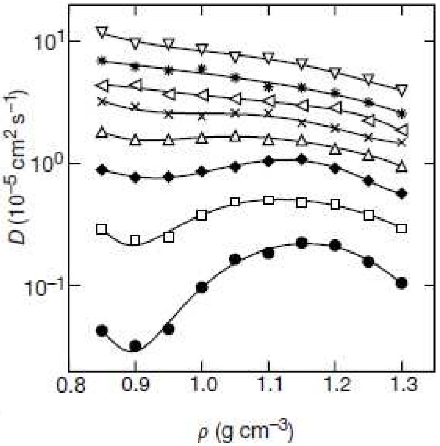


Bauchy et al. Chem Geol. 2013

- Species dependent. Na diffuses faster than Si or O in silicates

Behavior of the diffusion constant with other thermodynamic variables

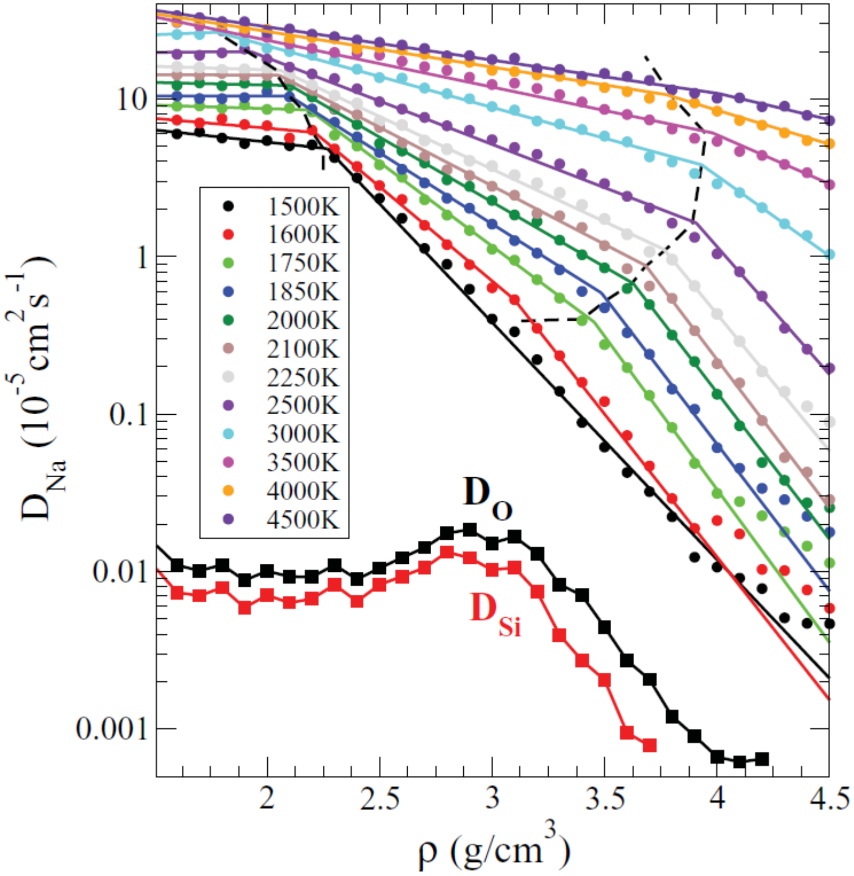
- Dependence on V or P.
- To be comparable with experiment, should be calculated at P=0.
- Usually at fixed V (e.g. V_g) and P non-zero



H_2O

Figure 3 Density dependence of the diffusion coefficient, D, shown for eight isotherms. The curves from top to bottom correspond to temperatures T (in K) of: 400, 350, 320, 300, 280, 260, 240 and 220. The solid lines are fifth-order polynomial fits to the data, and are simply guides to the eye. The diffusion coefficient was calculated from the long-time behaviour of the mean squared displacement of the water molecules, $\langle r^2(t)\rangle$, using the Einstein relationship $6D = d\langle r^2(t)\rangle/dt$.

Errington et al. Nature 2001



Bauchy et al., PRB 2011

- ☐ To go further in correlation study, one can follow correlation both in space and time. This information is richer, but the price to pay is the calculation of a two-variable correlation function, at least.
- ☐ We introduce these functions because they are also available in experiments, essentially form neutron scattering.

B) VAN HOVE CORRELATION FUNCTION

We first introduce a density correlation function G(**r**,**r’**,t) defined from the local atomic densities:

$$\rho G(\mathbf{r},\mathbf{r}';t)=\langle\sum_{i=1}^{N}\sum_{j=1}^{N}\delta(\mathbf{r}'+\mathbf{r}-\mathbf{r}_i(t))\delta(\mathbf{r}-\mathbf{r}_j(0))\rangle$$

or :

$$\rho G(\mathbf{r},\mathbf{r}';t)=\langle\rho(\mathbf{r}'+\mathbf{r},t)\rho(\mathbf{r}',0)\rangle$$

For an homogeneous system, G depends only on the relative distance. Integration gives:

$$G(\mathbf{r},t)=\frac{1}{N}\langle\sum_{i=1}^{N}\sum_{j=1}^{N}\ \delta(\mathbf{r}-\mathbf{r}_i(t)+\mathbf{r}_j(0))\rangle$$

$$G(\mathbf{r},t) = \frac{1}{N}\langle\sum_{i=1}^{N}\sum_{j=1}^{N} \delta(\mathbf{r} - \mathbf{r}_i(t) + \mathbf{r}_j(0))\rangle$$

At t=0, we have
$$G(\mathbf{r},0) = \frac{1}{N}\langle\sum_{i=1}^{N}\sum_{j=1}^{N}\delta(\mathbf{r} + \mathbf{r}_i(0) - \mathbf{r}_j(0))\rangle$$
$$= \delta(\mathbf{r}) + \rho g(\mathbf{r})$$

Except a singularity at the origin, the Van Hove correlation function is proportional to the pair correlation function g(r). We can split the function into two parts, **self and distinct**:

$$G(\mathbf{r},0) = G_s(\mathbf{r},0) + G_d(\mathbf{r},0)$$

which can be established at non-zero times:

$$G_s(\mathbf{r},t) = \frac{1}{N}\langle\sum_{i=1}^{N}\delta((\mathbf{r} + \mathbf{r}_i(0) - \mathbf{r}_i(t))\rangle$$

$$G_d(\mathbf{r},t) = \frac{1}{N}\langle\sum_{i\neq j}^{N}\delta(\mathbf{r} + \mathbf{r}_j(0) - \mathbf{r}_j(t))\rangle$$

Physical interpretation:

- The Van Hove function is the probability density of finding a particle i in the vicinity of r at time t, knowing that a particle j is in the vicinity of the origin at time t=0.
- The self part $G_s(r,t)$ is the probability density of finding a particle i at time t knowing that this particle was at the origin at time 0. Probability that a particle has moved a distance r in time t (dynamics).

$$G_s(\mathbf{r},t)=\frac{1}{N}\langle\sum_{i=1}^{N}\delta((\mathbf{r}+\mathbf{r}_i(0)-\mathbf{r}_i(t))\rangle$$

- The distinct part $G_d(r,t)$ is the probability density of finding a particle j different from i at time t knowing that the particle I was at the origin at time t=0. Probability to find at time t a different particle at a distance r from a place at which at time t=0 there was a particle. And $G_d(r,0)=g(r)$.

$$G_d(\mathbf{r},t)=\frac{1}{N}\langle\sum_{i\neq j}^{N}\delta(\mathbf{r}+\mathbf{r}_j(0)-\mathbf{r}_j(t))\rangle$$

Normalization of the functions leads to :

$$\int d\mathbf{r} G_s(\mathbf{r},t) = 1 \qquad \int d\mathbf{r} G_d(\mathbf{r},t) = N-1$$

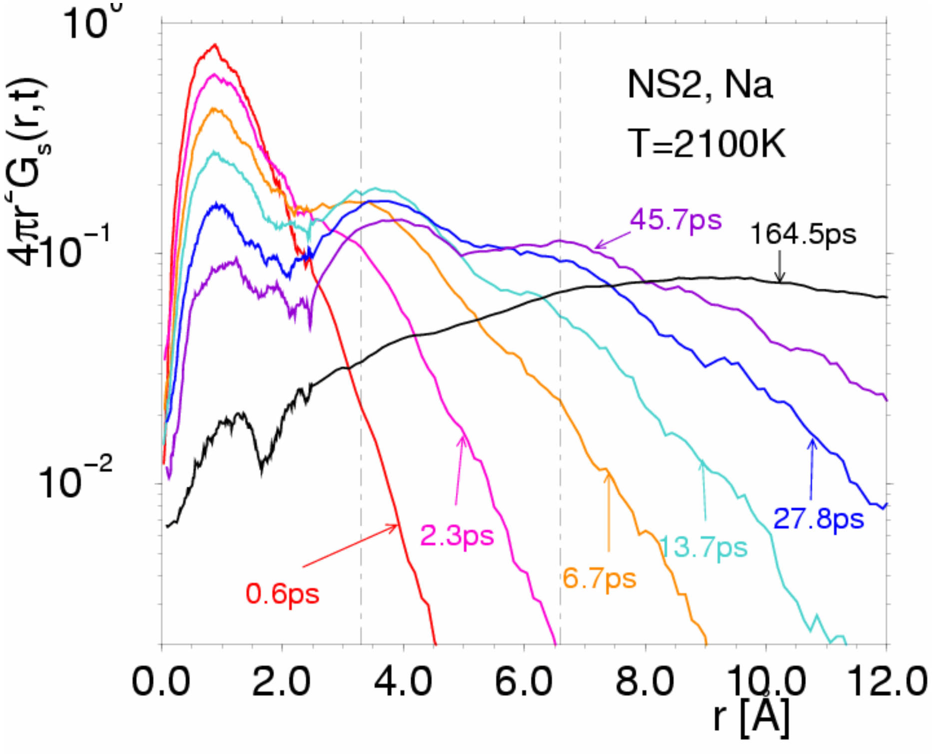
Integration over space, $G_d(r,t)$ is able to count the remaining particles

In the long-time limit, the system looses memory of the initial configuration and the correlation functions become independent of the distance:

$$\lim_{r\to\infty} G_s(\mathbf{r},t) = \lim_{t\to\infty} G_s(\mathbf{r},t) \simeq \frac{1}{V} \simeq 0$$

$$\lim_{r\to\infty} G_d(\mathbf{r},t) = \lim_{t\to\infty} G_s(\mathbf{r},t) \simeq \frac{N-1}{V} \simeq \rho$$

Examples: Self part in Na silicates



Kob, PRB 2000

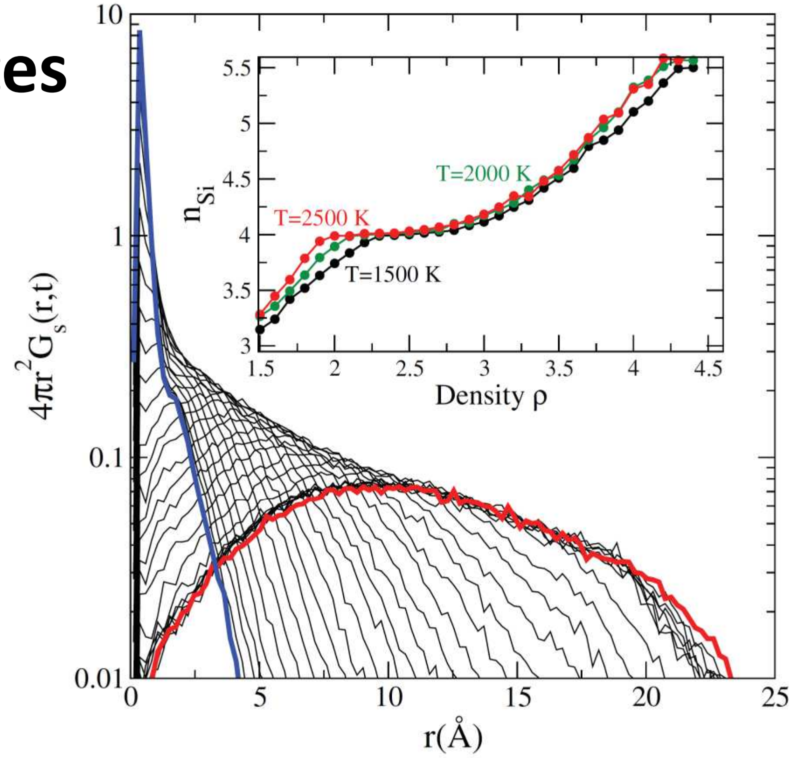


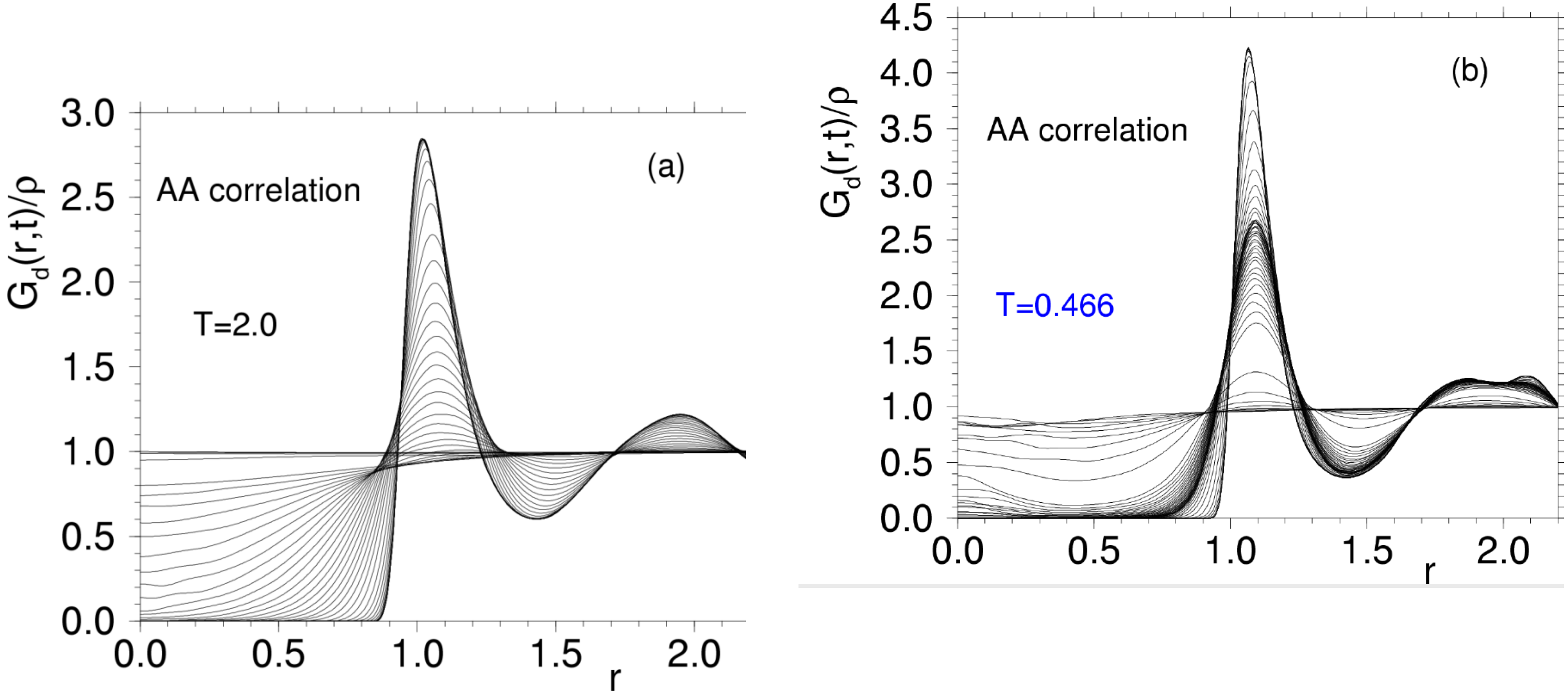
FIG. 4. (Color online) Van Hove function $G_s(r,t)$ of the NS3 system at 2000 K and fixed time $t = t_0 = 20$ ps for various densities between 2.0 and 4.5 g/cm^3. The red and blue curves correspond to 2.0 and 4.5 g/cm^3, respectively. The insert shows the change in Si coordination number n_{Si} as a function of density for three selected temperatures.

Bauchy, PRB 2011

- At short times, $G_s(r,t) \sim \delta(r)$
- Small times: rattling and hopping motion on the length scale of nearest neighbors.
- Spatial extent of the motion is determined by thermodynamic conditions: T (viscous slowing down) or P (motion blocked by the high density).

Examples: Distinct part in a binary A-B mixture

- ☐ At high temperature, the correlation hole at r=0 is quickly filled up
- ☐ At low temperature, correlation hole survives at long time

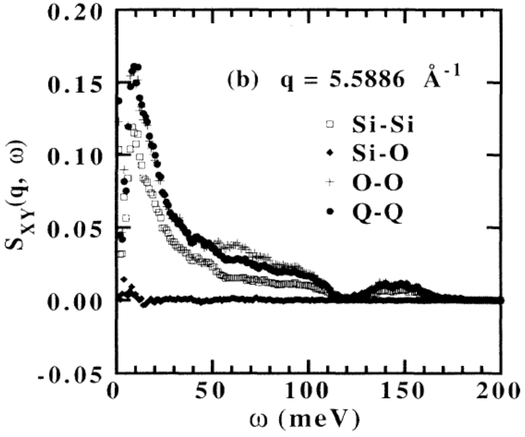


Kob and Andersen, PRE 1995

C) DYNAMIC STRUCTURE FACTOR

Fourier (time) transform of the intermediate scattering function (see next lectures)

$$S(\mathbf{k},\omega) = \int dt F(\mathbf{k},t)e^{i\omega t} \qquad \int d\omega S(\mathbf{k},\omega) = S(\mathbf{k})$$

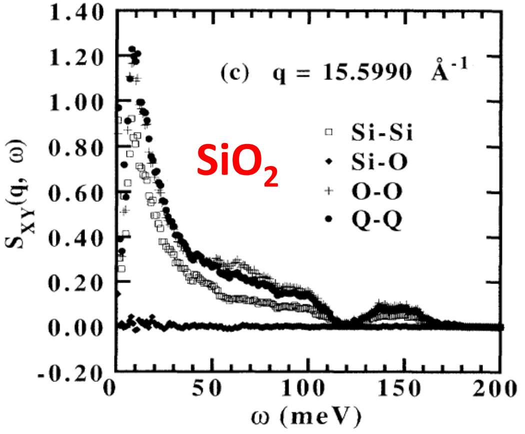


Neutron weighted dynamic structure factor
(similar to static one):

$$S_n(\mathbf{q},\omega) = \frac{1}{N\langle b^2\rangle}\int_{-\infty}^{\infty}\frac{dt}{2\pi}e^{-i\omega t}\sum_{i=1}^{N}\sum_{j=1}^{N} b_i b_j \langle e^{i\mathbf{q}\cdot\mathbf{r}_j(t)}e^{-i\mathbf{q}\cdot\mathbf{r}_i(0)}\rangle$$

Access to partial dynamic structure factors :

$$S_{\alpha\beta}(\mathbf{q},\omega) = \frac{1}{\sqrt{N_\alpha N_\beta}}\int_{-\infty}^{\infty}\frac{dt}{2\pi}e^{-i\omega t}\sum_{i\subset\alpha}^{N_\alpha}\sum_{j\subset\beta}^{N_\beta}\langle e^{i\mathbf{q}\cdot\mathbf{r}_j(t)}e^{-i\mathbf{q}\cdot\mathbf{r}_i(0)}\rangle$$



SiO_2

- ☐ Can be compared to measurements extracted from coherent inelastic neutron scattering.

Jin and Vashishta PRB 1993

Differential cross-section ($d^2\sigma/d\omega d\Omega$) proportional to S(q,ω)

D) LINEAR RESPONSE THEORY

❑ Goal: Having MD generated trajectories at our disposal, we want to compute
- viscosity, electrical or thermal conductivity, mechanical properties, etc.

Use of linear response theory

❑ General idea (Onsager): Disturbance in a system created by a weak external perturbation decays in the same way as a spontaneous fluctuation in equilibrium.

❑ Linear response theory : link between time correlation functions and response to weak perturbations (Green-Kubo's fluctuation-dissipation relations)

❑ **Static response** (e.g. polarization, electric current) to a constant weak applied field.

- Property characterized by a dynamic variable A.
- Change in the Hamiltonian $H_0-\lambda B(\mathbf{p}^N,\mathbf{q}^N)$
- External perturbation brings the system from Ensemble average $\langle A\rangle_0$ to $\langle A\rangle_0+\langle\Delta A\rangle$.

- External perturbation brings the system from Ensemble average $<A>_0$ to $<A>_0+<\Delta A>$.

- The Ensemble average <A> for this perturbated A is :

$$\langle A\rangle_0 + \langle \Delta A\rangle = \frac{\int d\Gamma\ \exp[-\beta(\mathcal{H}_0 - \lambda B)]A}{\int d\Gamma\ \exp[-\beta(\mathcal{H}_0 - \lambda B)]}$$

Γ is the integration over phase space variables ($\mathbf{q}^N$,$\mathbf{p}^N$)

- <A> can be expressed as a linear Taylor expansion: $<A>=<A>_0+\lambda(d<A>/d\lambda)_{\lambda=0}$ involving the quantity:

$$\left(\frac{\partial\langle\Delta A\rangle}{\partial\lambda}\right)_{\lambda=0} = \beta\{\langle AB\rangle_0 - \langle A\rangle_0\langle B\rangle_0\}$$

❑ Similarly, we can compute the **<u>dynamic response</u>**

- ❖ Preparation of the system under a weak constant perturbation. One thus has $<A>=<A>_0+<\Delta A>$
- ❖ At t=0, the perturbation is switched off. Reponse ΔA decays to zero.
- ❖ Ensemble average of ΔA at time t is given by:

$$\langle \Delta A(t)\rangle = \frac{\int d\Gamma\ \exp[-\beta(\mathcal{H}_0 - \lambda B)]A(t)}{\int d\Gamma\ \exp[-\beta(\mathcal{H}_0 - \lambda B)]}$$

with A(t) the value of A at time t in a system starting at point Γ.

$$\langle \Delta A(t) \rangle = \frac{\int d\Gamma \; \exp[-\beta(\mathcal{H}_0 - \lambda B)] A(t)}{\int d\Gamma \; \exp[-\beta(\mathcal{H}_0 - \lambda B)]}$$

In the limit λ=0 (and assuming that $\langle A \rangle_0 = 0$), we can simplify to:

$$\begin{aligned} \langle \Delta A(t) \rangle &= \beta\lambda \frac{\int d\Gamma \; \exp[-\beta \mathcal{H}_0] B A(t)}{\int d\Gamma \; \exp[-\beta \mathcal{H}_0]} \\ &= \beta\lambda \langle B(0) A(t) \rangle \, . \end{aligned}$$

The decay of ΔA of the system is determined by a time correlation function describing the decay of spontaneous fluctuations of A in equilibrium under an external perturbation B.

E) TIME CORRELATION FUNCTION

- Let us consider two time dependent signals A and B. The general time correlation between them is given by:

$$C(t) = \lim_{\tau \to \infty} \frac{1}{\tau} \int_0^{\tau} A(t_0) B(t_0 + t) dt_0 = \left\langle A(t_0) B(t_0 + t) \right\rangle$$

A=B: auto-correlation. Otherwise cross-correlation which is the case in linear response

- At equilibrium, the correlation is invariant under time translation, i.e.

$$C(t) = \langle A(t_0) B(t_0 + t) \rangle = \langle A(t_0 + s) B(t_0 + s + t) \rangle$$

NB: Glasses C(t) is not time invariant so that t depends on $t_0 = t_w$ (waiting time)

- In the limit of no delay time, C(0) is the static correlation function

- Can be normalized to the value at t_0:

$$C(0) = \lim_{\tau \to \infty} \frac{1}{\tau} \int_0^{\tau} A(t_0) B(t_0)\, dt_0$$

$$= \langle A(t_0) B(t_0) \rangle \equiv \langle AB \rangle$$

$$\hat{C}(t) = \frac{\langle A(t_0) B(t_0 + t) \rangle}{\langle AB \rangle}$$

G) APPLICATIONS-1: ELECTRICAL CONDUCTIVITY

Hamiltonian of the system with vector potential is given by $\mathcal{H}' = \sum_{i=1}^{N} \frac{1}{2m_i}\left(\mathbf{p}_i - \frac{e_i}{c}\mathbf{A}\right)^2 + \mathcal{U}_{\text{pot}}.$

A being a jauge field.
I remind that from in Classical Electromagnetism, the generalized momentum p of a particle with mass m and charge q moving at a velocity v in a vector potential A is $\mathbf{p}=m\mathbf{v}+e_i\mathbf{A}$.

- ❑ The vector potential is switched off at t=0. The electrical field is an infinitesimal spike (delta function) given that:

$$\mathbf{E} = -\frac{1}{c}\dot{\mathbf{A}} \qquad \mathbf{E}(t) = \frac{1}{c}\mathbf{A}\delta(t)$$

- ❑ At the first order, we can write :

$$\begin{aligned}
\mathcal{H}' &= \mathcal{H}_0 - \sum_{i=1}^{N} \frac{e_i}{cm_i}\mathbf{p}_i \cdot \mathbf{A} + \mathcal{O}(A^2) \\
&= \mathcal{H}_0 - \frac{\mathbf{A}}{c}\int d\mathbf{r} \sum_{i=1}^{N} \frac{e_i}{m_i}\mathbf{p}_i \delta(\mathbf{r}_i - \mathbf{r}) \\
&= \mathcal{H}_0 - \frac{\mathbf{A}}{c}\int d\mathbf{r}\, \mathbf{j}(\mathbf{r}),
\end{aligned}$$

with **j**(**r**) the current density

From the expression of the perturbated Hamiltonian, we obtain the correlation function:
$$\mathcal{H}' = \mathcal{H}_0 - \frac{\mathbf{A}}{c}\int d\mathbf{r}\, \mathbf{j}(\mathbf{r})$$

we can write the average current density as:
$$\langle \boldsymbol{j}(t)\rangle = \frac{\int d\Gamma e^{-\beta\left(H_0 - \frac{A}{c}\int dr' \boldsymbol{j}(r',t)\right)} \boldsymbol{j}(\boldsymbol{r})}{\int d\Gamma e^{-\beta H_0}}$$

and, expanding at small vector A, we have:
$$\langle \mathbf{j}(t)\rangle = \frac{\mathbf{A}}{cVk_BT}\int d\mathbf{r}d\mathbf{r}'\, \langle \mathbf{j}(\mathbf{r},0)\mathbf{j}(\mathbf{r}',t)\rangle$$

Remember that we have Ohm's law (**j**=σ**E**) and for the response function :
$$\langle \Delta A(t)\rangle = \int_{-\infty}^{t} dt' \chi_{AB}(t-t')f(t')$$

so that (E(t) is a Dirac function):
$$\begin{aligned}\langle \mathbf{j}(t)\rangle &= \int_{-\infty}^{t} dt' \sigma(t-t')E(t') \\ &= \sigma(t)\frac{A}{c}.\end{aligned}$$

and finally:
$$\boxed{\sigma(t) = \frac{1}{Vk_BT}\int d\mathbf{r}d\mathbf{r}'\, \langle \mathbf{j}(\mathbf{r},0)\mathbf{j}(\mathbf{r}',t)\rangle}$$

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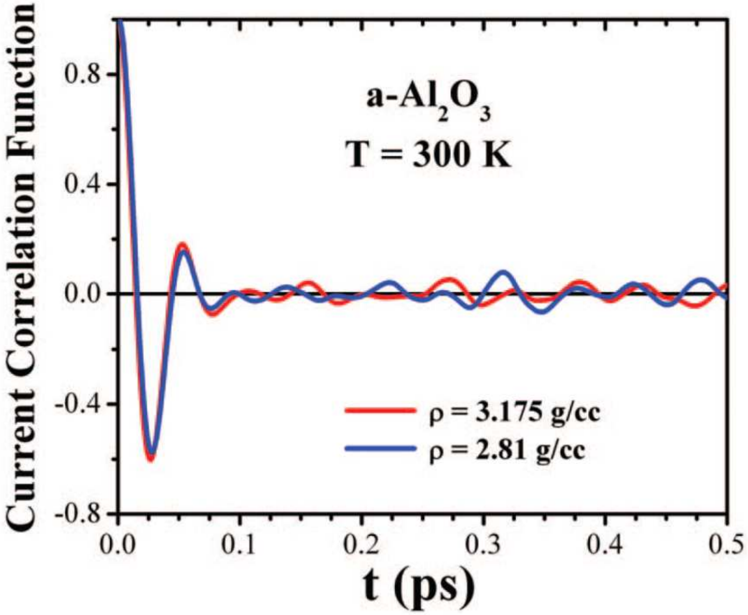
Example-1: Electrical conductivity in amorphous aluminia

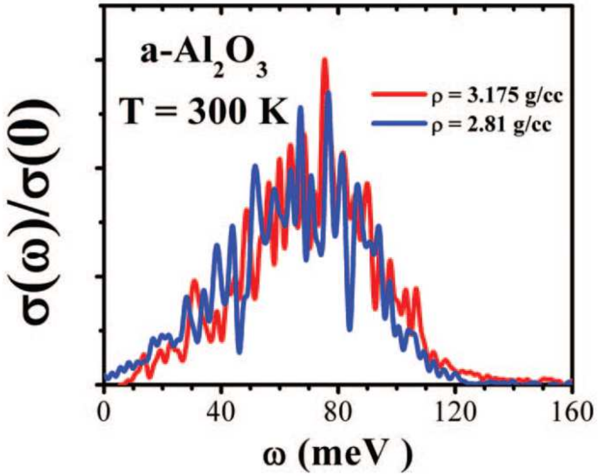
Computation of the current from the atomic velocities: $$\mathbf{J}(t) = \sum_i Z_i e \mathbf{v}_i(t)$$

Normalized current autocorrelation function: $$\psi(t) = \frac{\langle \mathbf{J}(t) \cdot \mathbf{J}(0) \rangle}{\langle \mathbf{J}(0) \cdot \mathbf{J}(0) \rangle}$$



Frequency dependent conductivity

$$\sigma(\omega) = \frac{\langle J(0)^2 \rangle}{3Vk_BT} \int_0^\infty \psi(t) e^{i\omega t} dt$$



Vashishta et al. JAP 2008

Example-2: Electrical conductivity in an aluminosilicate liquid

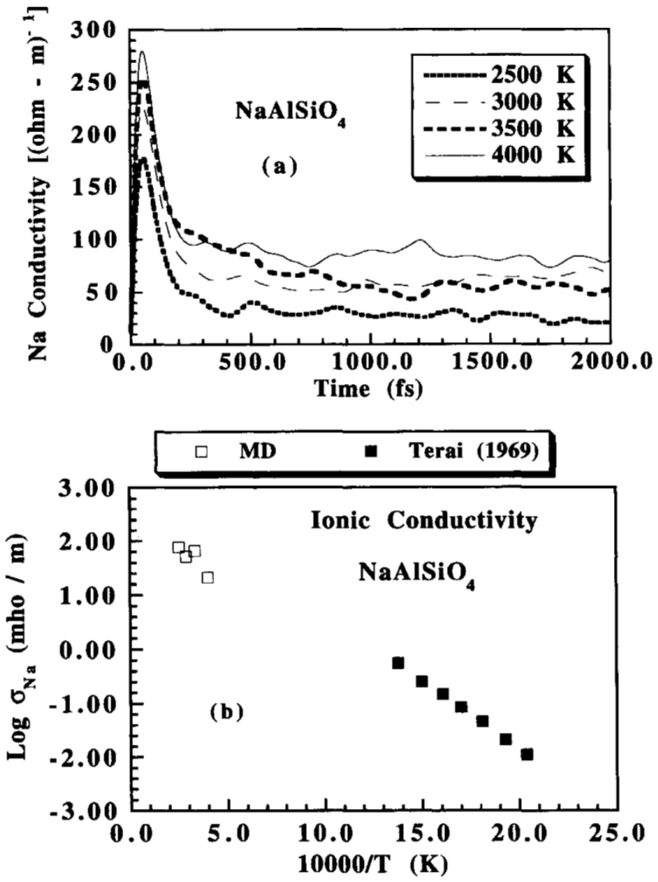


FIGURE 4. **(a)** Ionic conductivity due to Na in $NaAlSiO_4$ determined from simulations at four temperatures, calculated according to Equations 7 and 8 in text. **(b)** Ionic conductivity determined in simulations (open squares) in comparison with results of laboratory measurements by Terai (1969).

Spera et al. JCP 2001

- Plot of $\sigma(t)$ following:

$$\sigma(t) = \frac{1}{Vk_BT} \int d\mathbf{r}d\mathbf{r}' \langle \mathbf{j}(\mathbf{r},0)\mathbf{j}(\mathbf{r}',t)\rangle$$

- Access to σ_{dc}

$$\sigma(\omega = 0) = \frac{1}{Vk_BT} \int_0^\infty dt \int d\mathbf{r}d\mathbf{r}' \langle \mathbf{j}(\mathbf{r},0)\mathbf{j}(\mathbf{r}',t)\rangle$$

- Behavior with temperature (Arrhenius)

Example-3: Beyond linear response theory-usefulness of MD

- The computation of $\sigma(t)$ or σ_{dc} and successful comparison with experiment is the starting point (prerequisite) of atomic scale insight of conductivity.

- Channel conduction in silicates

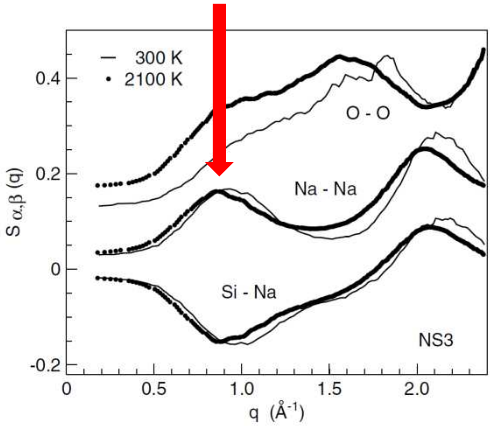
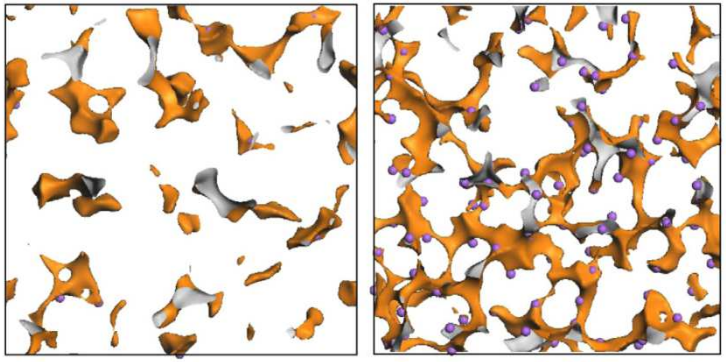


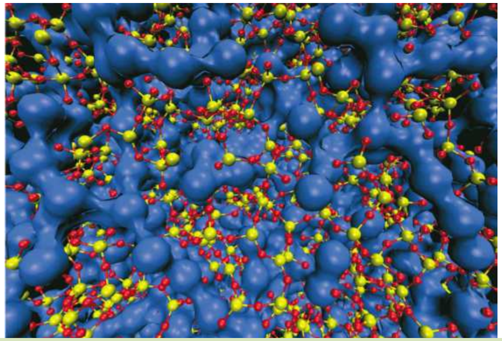
FIG. 3. Partial structure factors $S_{O,O}(q)$, $S_{Na,Na}(q)$, $S_{Si,Na}(q)$ as obtained by the simulation in glassy and liquid sodium trisilicate at experimental densities. $S_{O,O}(q)+0.1$ for clarity.



ces of the isosurfaces of constant mobile ion energy with regions of low site energies in the glasses $xNa_2O-(1-x)SiO_2$ for $x=0.1, 0.45$ (Na: spheres) (left to right), projected z-axis (thickness nearly 5 Å).

Prasada Rao et al. SS Ionics 2011

Link with static structure factor
Typical distance related to
q=0.9A^{-1}



Meyer et al. PRL 2004

F) APPLICATIONS-2: THERMAL CONDUCTIVITY

Similar treatment to electrical conductivity.

☐ Define a heat current

$$\mathbf{J} = \sum_i E_i \mathbf{v}_i + \frac{1}{2} \sum_{\substack{i>j \\ pairs}} \mathbf{r}_{ij}[\mathbf{F}_{ij} \cdot (\mathbf{v}_i + \mathbf{v}_j)]$$

with E_i, energy of atom i

☐ Compute the autocorrelation function:

$$\kappa = \frac{1}{3Vk_BT^2} \int_0^\infty < \mathbf{J}(t) \cdot \mathbf{J}(0) > dt$$

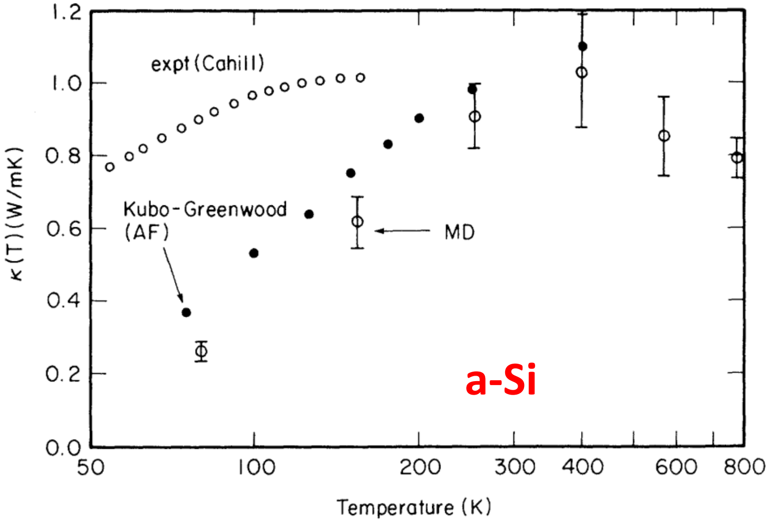
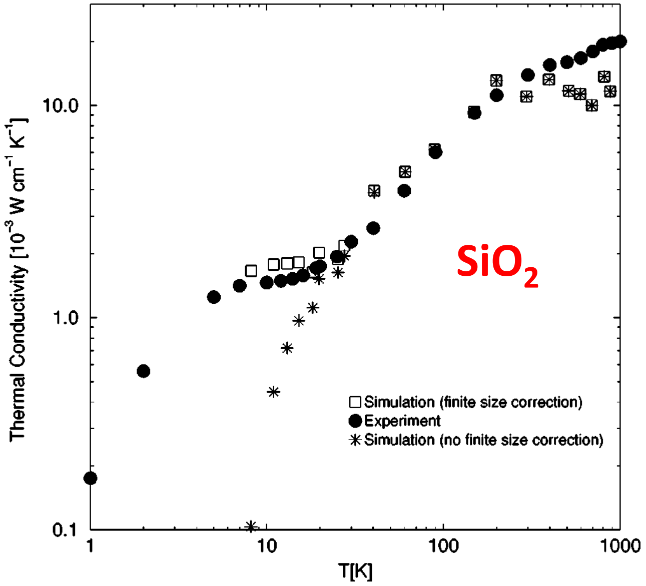


FIG. 4. Thermal conductivity vs temperature for amorphous silicon. Solid circles are from Allen and Feldman (Ref. 5) and open circles are from experiment (Ref. 17). Open circles with error bars are the present calculations.

Lee et al., PRB 1991



Jund and Jullien, PRB 1999

F) APPLICATIONS-3: VISCOSITY

- Shear can not be interpreted in terms of an external field acting on all particles.

- Use of a canonical transformation corresponding to uniform shear transforming linearly the coordinates from $\mathbf{r}^N$ to $\mathbf{r}'^N$ with $\mathbf{r}'_i = h_{ij}\mathbf{r}_j$ and $h_{ij} = 1+\varepsilon_{ij} \sim 1$

- This means that we transform the Hamiltonian from :

$$\mathcal{H}_0 = \sum_{i=1}^{N} p_i^2/(2m_i) + \mathcal{U}(\mathbf{r}^N)$$

to:

$$\mathcal{H}_1 = \sum_{i=1}^{N} \frac{1}{2m_i} \mathbf{p}'_i \cdot \mathsf{G}^{-1} \cdot \mathbf{p}'_i + \mathcal{U}(\mathbf{r}'^N)$$

with G the metric tensor defined as: $\mathsf{G} \equiv \mathsf{h}^T \cdot \mathsf{h}$

- Effect of uniform shear, e.g. $\varepsilon_{xy}=\varepsilon$ otherwise 0.

- Assume equilibration of the system with Hamiltonian H_1 (i.e. under shear) and then switch off ε at t=0 (now left with H_0).

- ☐ Assume equilibration of the system with Hamiltonian H_1 (i.e. under shear) and then switch off ε at t=0 (now left with H_0).

- ☐ The system experiences a δ-function spike in the shear rate, i.e. one has:

$$\frac{\partial v_x}{\partial y} = -\epsilon\delta(t)$$

- ☐ Time-dependent response of the shear stress, $\sigma_{xy}(t)$ to the sudden change from H_1 to H_0:

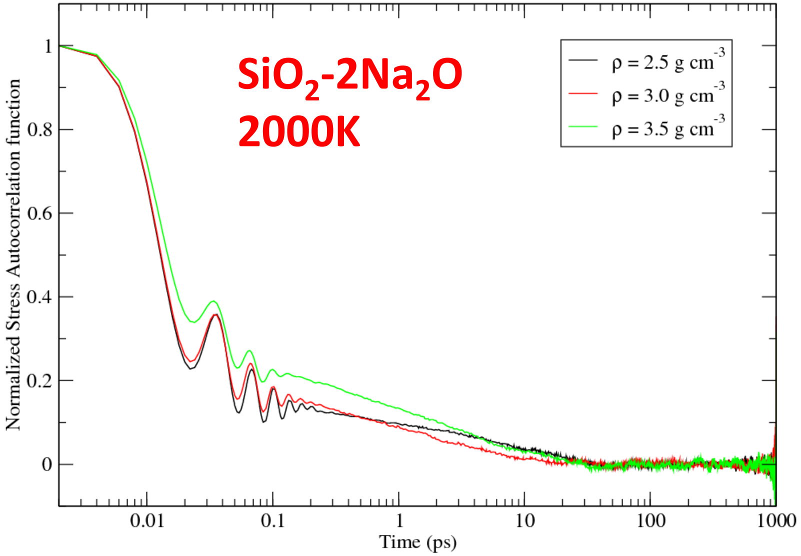
$$\langle \sigma_{xy}(t)\rangle = -\epsilon \frac{1}{Vk_BT}\langle \sigma_{xy}(0)\sigma_{xy}(t)\rangle$$

where σ_{xy} (tensor) can be computed via:

$$\sigma_{\alpha\beta} = \frac{1}{V}\sum_{i=1}^{N}\left[\frac{(\mathbf{p}_i\cdot\hat{\mathbf{e}}_\alpha)(\mathbf{p}_i\cdot\hat{\mathbf{e}}_\beta)}{m_i} + (\mathbf{q}_i\cdot\hat{\mathbf{e}}_\alpha)(\mathbf{F}_i\cdot\hat{\mathbf{e}}_\beta)\right]$$

Remember (lecture 5) that pressure is :

$$p = \frac{1}{3V}\sum_{i=1}^{N}\left[\frac{\mathbf{p}_i^2}{m_i} + \mathbf{q}_i\cdot\mathbf{F}_i\right]$$



- Again, remember that: $\langle \Delta A(t) \rangle = \int_{-\infty}^{t} dt' \chi_{AB}(t-t') f(t')$

with f(t’) the δ-function of the shear rate.

We find that

$$\sigma_{xy} = \frac{\partial v_x}{\partial y} \times \frac{1}{V k_B T} \int_0^{\infty} dt \, \langle \sigma_{xy}(0) \sigma_{xy}(t) \rangle$$

- Then, we remember Newton’s law for viscosity.

Under laminar flow, the force exterted by a fluid in the x-direction is proportional to the velocity gradient, the constant of proportionality being the shear viscosity.

- The shear stress is thus : $\boldsymbol{\sigma_{xy} = \eta \frac{\partial v_x}{\partial y}}$ and, by identification:

$$\boxed{\eta = \frac{1}{V k_B T} \int_0^{\infty} dt \, \langle \sigma_{xy}(0) \sigma_{xy}(t) \rangle}$$

G) APPLICATIONS-3: VISCOSITY

Example-1: Viscosity of a silicate liquid (NS2) and MORB under pressure

NPT (P=0), then NVE

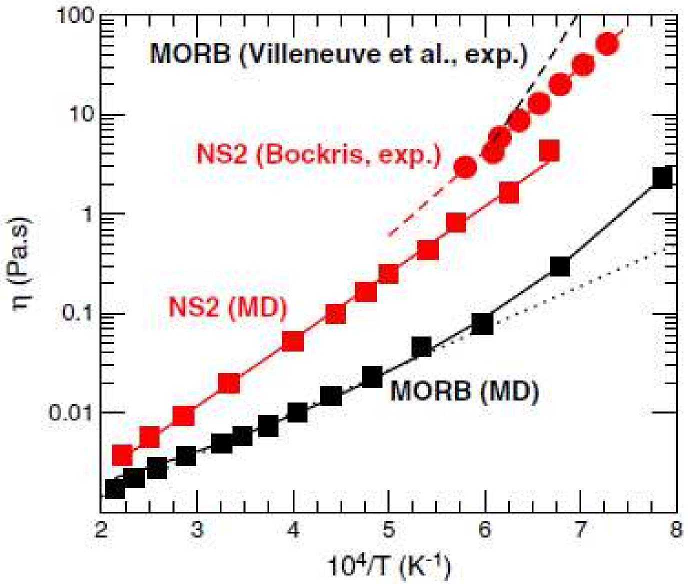
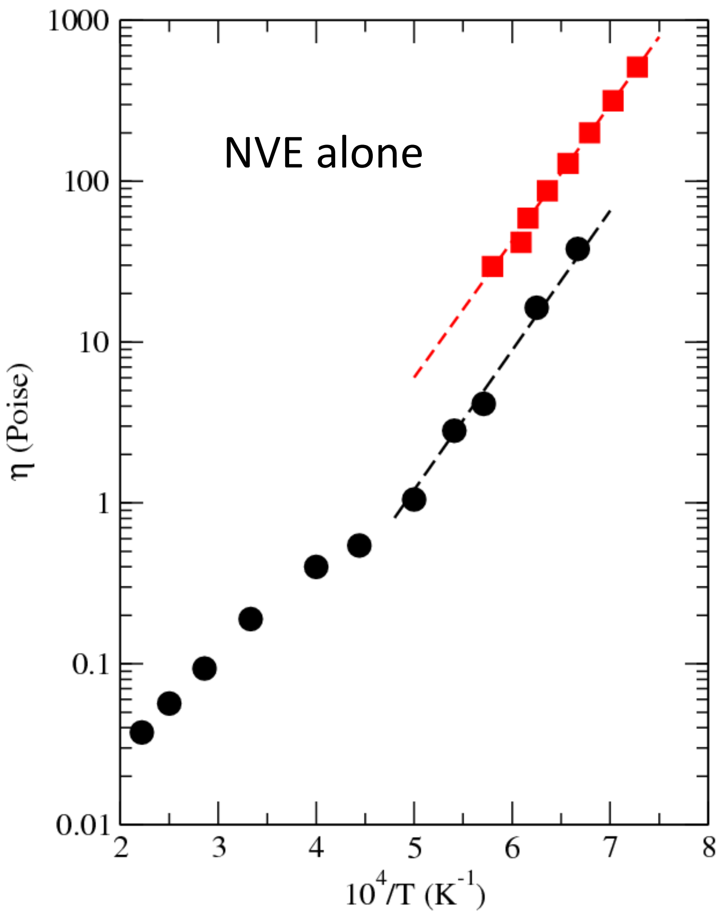


Fig. 3. Simulated viscosity of the NS2 liquid at zero pressure (filled red squares), compared to experimental data (red circles) from Bockris et al. (1955), together with simulated viscosity of the MORB liquid at zero pressure (filled black squares), compared to experimental data (broken black curve) from Villeneuve et al. (2008). The dotted line is a high temperature Arrhenius fit for the MORB data whereas the solid line is a TVF fit (see text for parameters).



Bauchy et al. Chem. Geol. 2013

G) APPLICATIONS-3: VISCOSITY

Example-1: Viscosity of a silicate liquid (NS2) and MORB under pressure

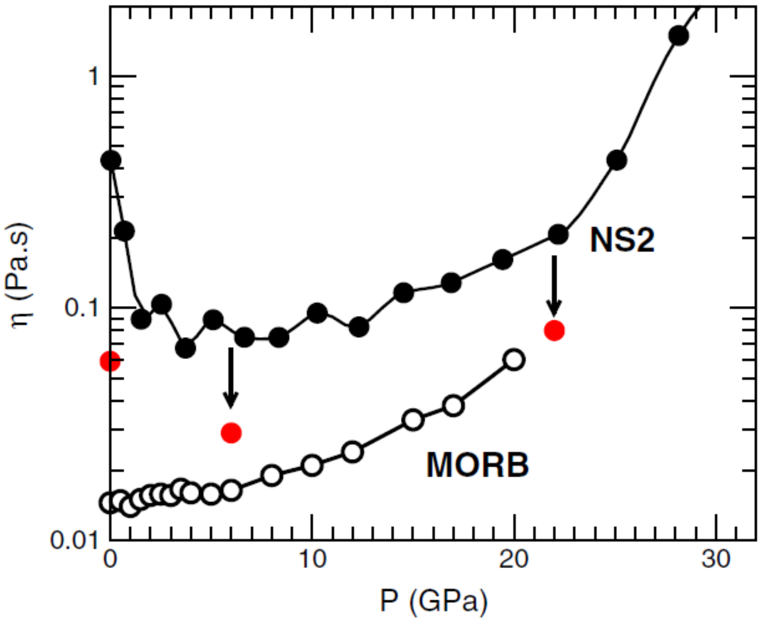


Fig. 6. Simulated viscosity of a NS2 liquid at T = 2000 K (filled circles) and a MORB liquid at T = 2273 K (open circles) as a function of pressure. Red circles correspond to the simulated viscosity of three selected densified NS2 using the potential of Guillot and Sator (2007a).

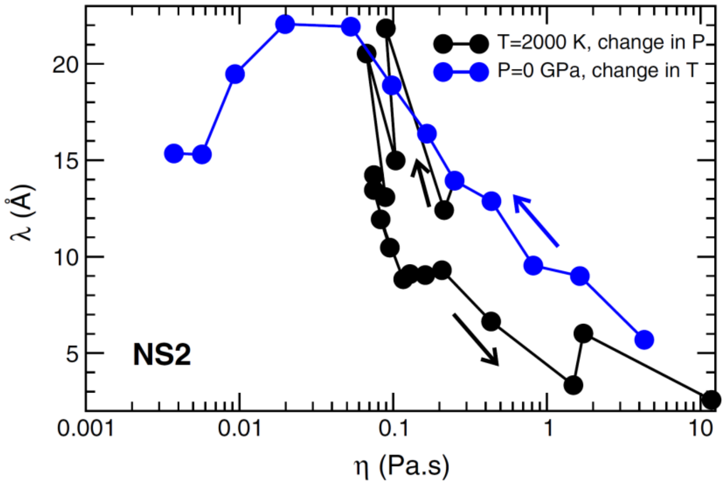
❑ Detecting anomalies (minima in viscosity)

❑ Checking for empirical relationships

Eyring (1948): $\eta = k_B T/\lambda D$

With λ a jump distance ($d_{O\text{-}O}$~ a few A)

- Compute (MD) D and η
- Valid only at high viscosity



Example-2: Viscosity of silica

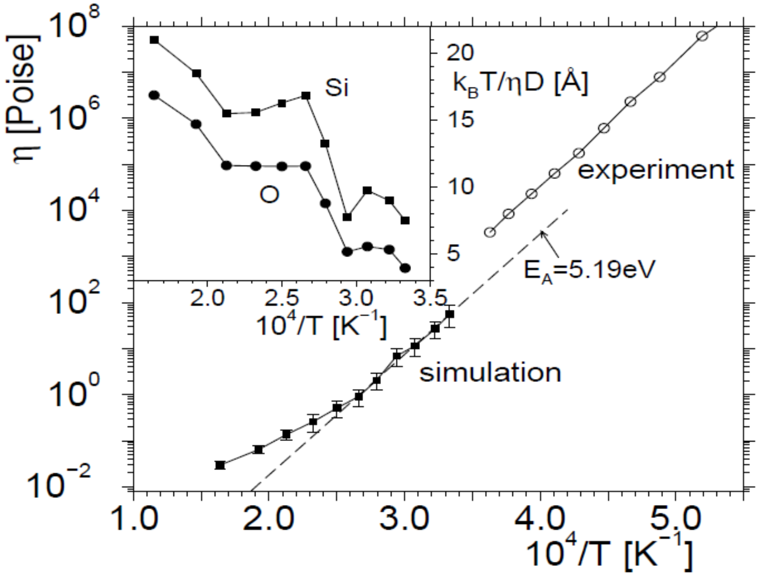


FIG. 10. Main figure: Arrhenius plot of the shear viscosity from the simulation (filled squares). The dashed line is a fit with an Arrhenius law to our low-temperature data. The open circles are experimental data from Urbain *et al.* [34]. Inset: Temperature dependence of the left hand side of Eq. (12) to check the validity of the Stokes-Einstein relation.

Horbach and Kob, PRB 1999

Conclusion:

☐ Dynamic quantities can be estimated from Molecular Dynamics
    Time-dependent structural correlations
    Transport coefficients (linear response theory)

☐ Insight into the glass transition phenomenon (see next lectures)

☐ Agreement with experiments is less obvious (as compared to structure)

Next time: Force fields and limitations

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