

# Glassiness in insulating granular Al thin films

Thierry Grenet, Julien Delahaye,  
Frédéric Gay, M. C. Cheynet\*,  
Maher Sabra, Jean Honoré,

*Institut Néel, CNRS Grenoble*

*\*SIMAP, Grenoble*



# OUTLINE

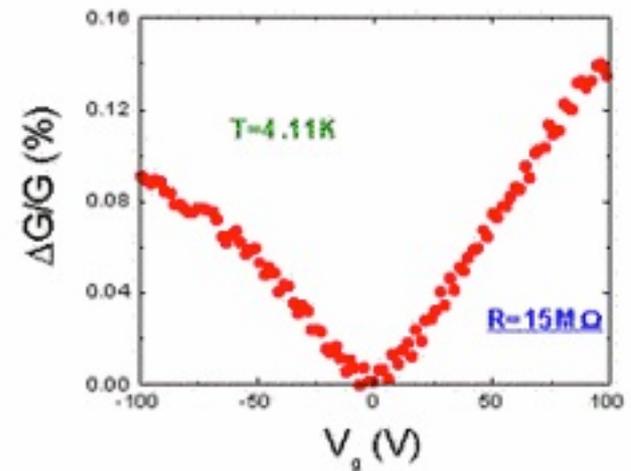
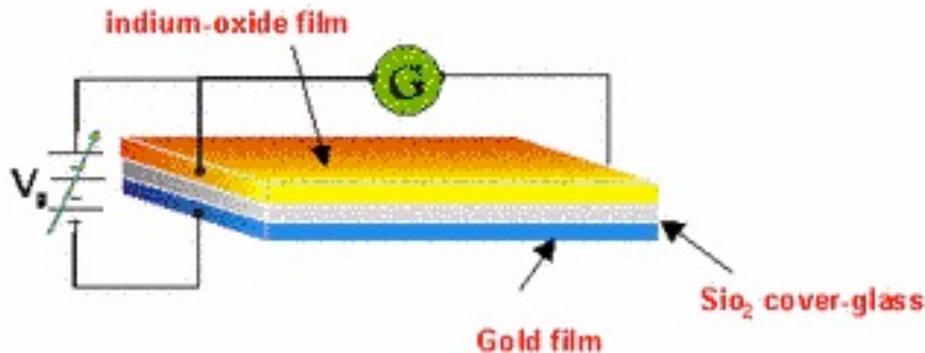
- how it started
- manifestations of « glassiness » in granular AI
- ageing
- questions

# Electron Coulomb glass ?

J. H. Davies, P. A. Lee and T. Rice (1982):

*localized electrons + unscreened coulomb repulsion → highly correlated → new glass (finite T glass transition?)*

**Ben Chorin et Ovadyahu (1991): anomalous field effect and very slow relaxation of conductance in insulating indium oxide**



**Manifestation of the electron (Coulomb ?) glass in indium oxide ...**

# Indium oxide ... what else ?

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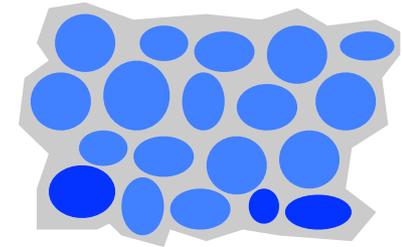
## QUESTIONS:

- *What is special with indium oxide ? Why no other system ?*

- *Standard doped semi-conductors:  $\emptyset$*

- *What about granular metals ?*

→ *look for these effects in granular Al*



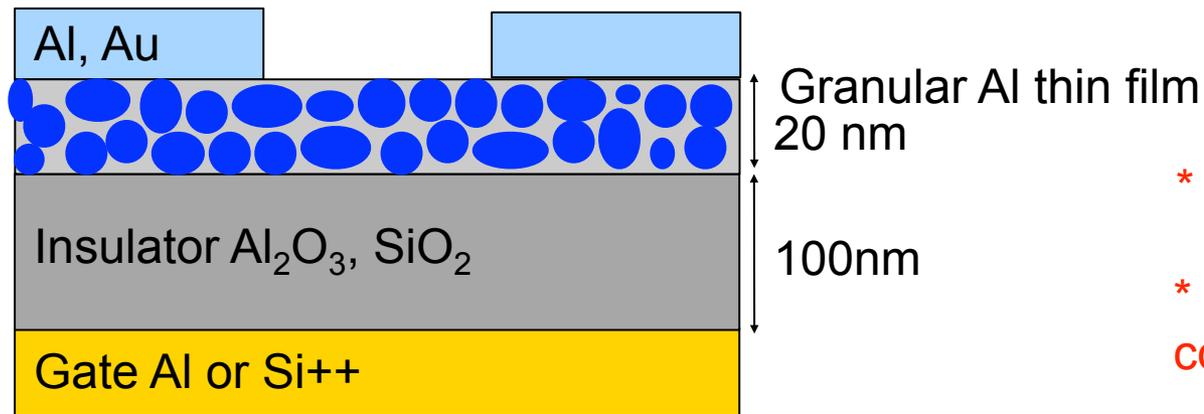
*actually seen in: granular gold (Adkins et al., 1984)*

*ultrathin lead (Goldman et al. , 1997 and 2001)*

# OUTLINE

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- manifestation of « glassiness » in granular AI
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# Granular Aluminium samples

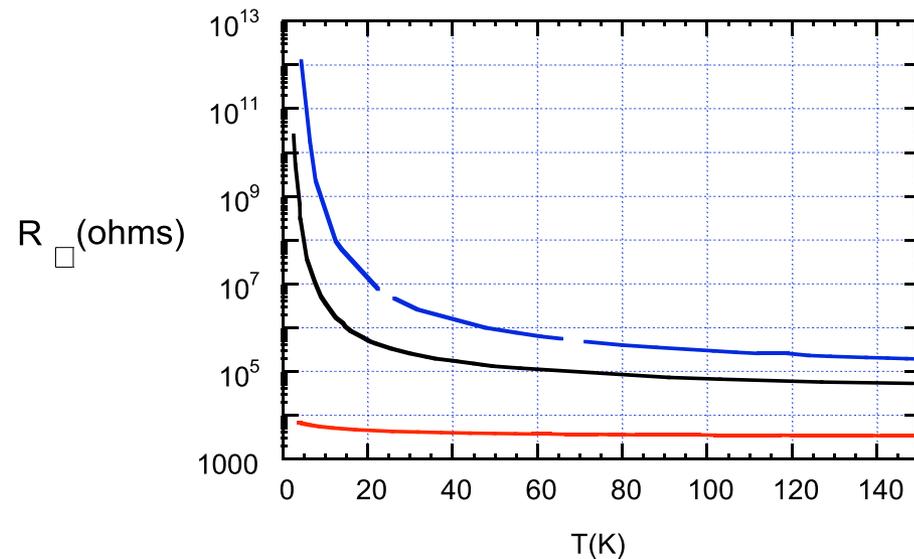


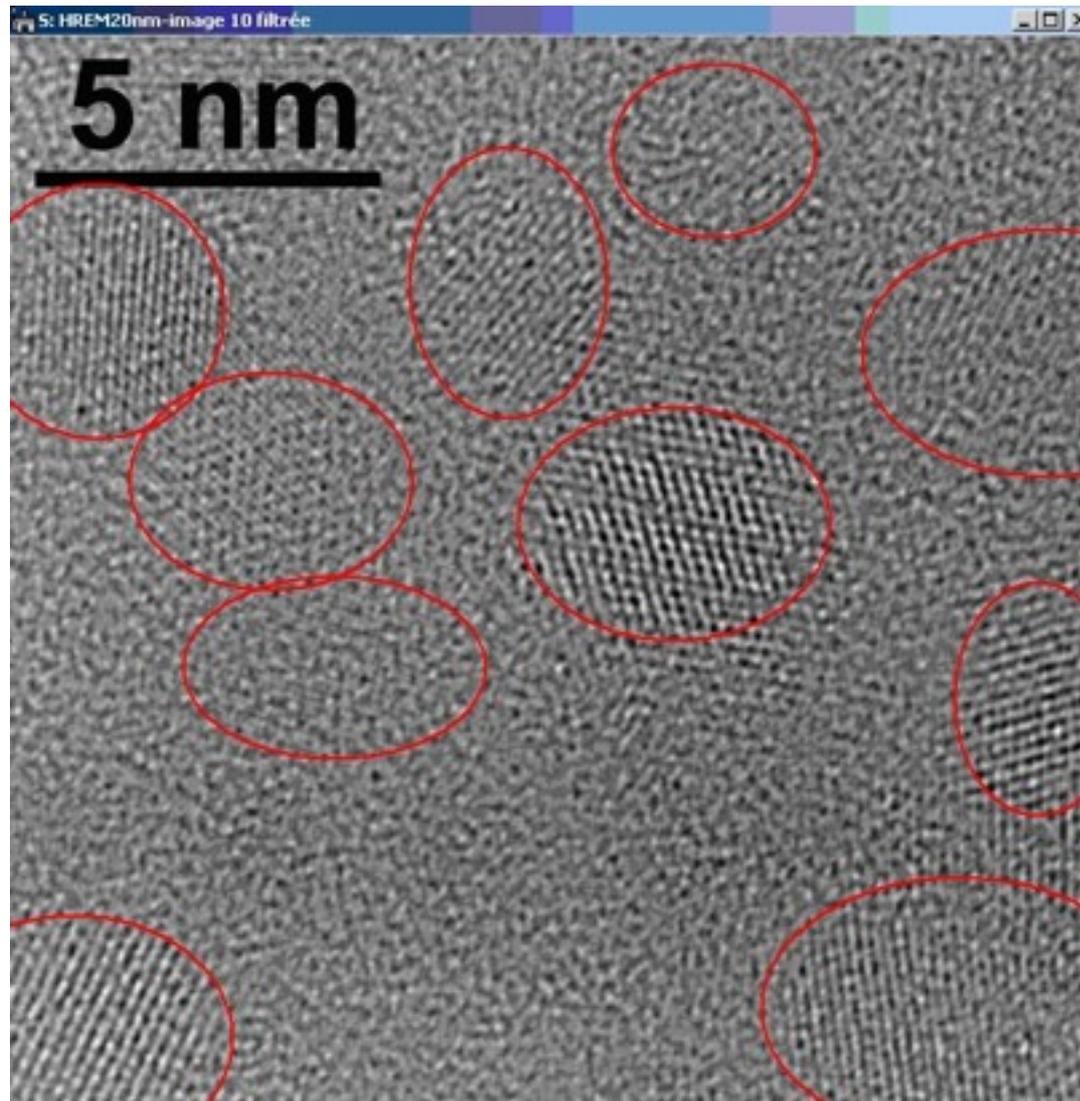
\* Al evaporated in P(O<sub>2</sub>)

\* nanometric Al grains covered by Al<sub>2</sub>O<sub>3</sub>

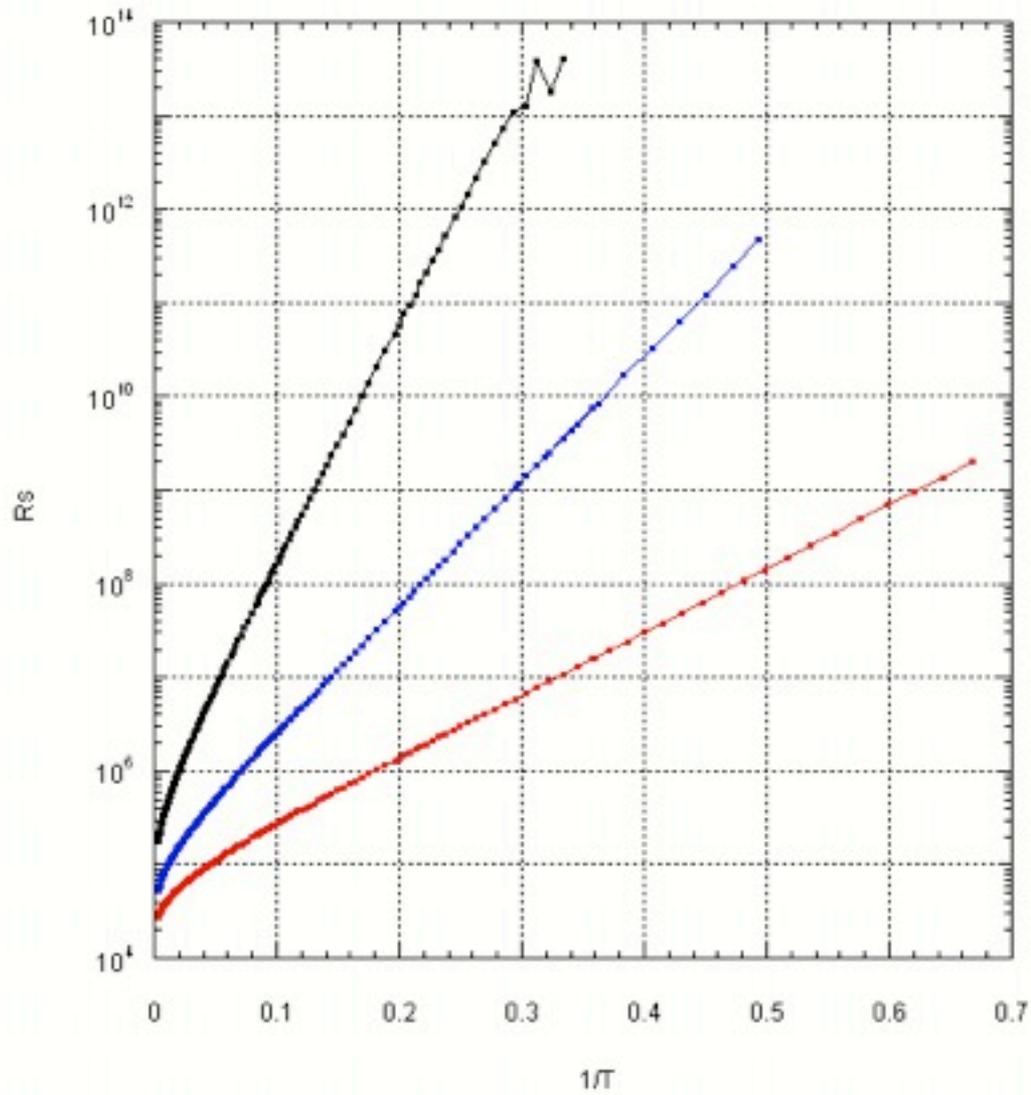
Study insulating films:

$R/\square$  at 4K: 100 k $\Omega$   $\rightarrow$  100 G $\Omega$



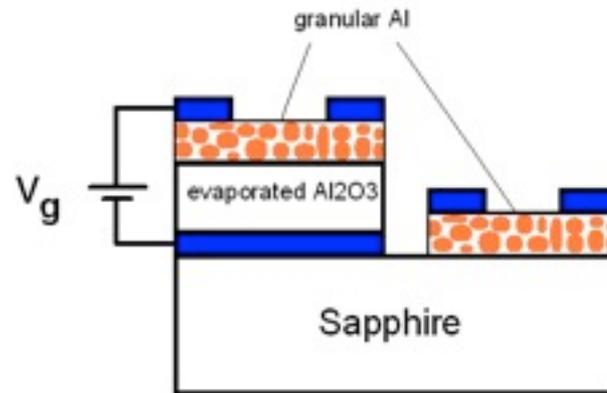
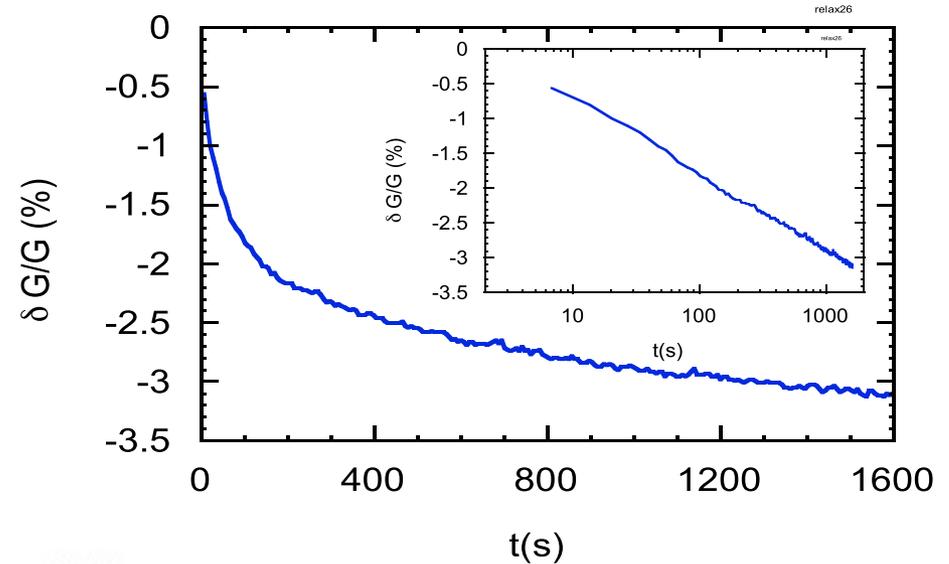
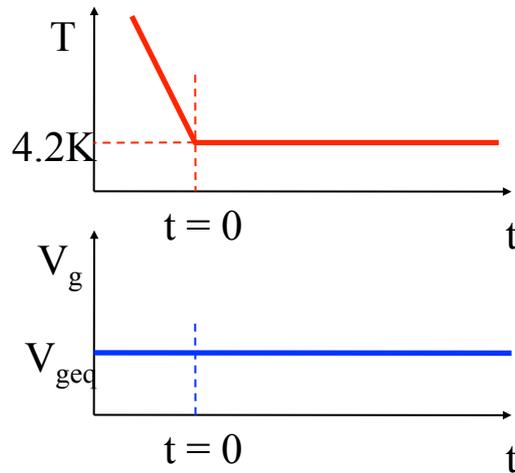


RT10nm2  
RT10NM21  
RT10nm13



# Out of equilibrium effects: slow relaxation

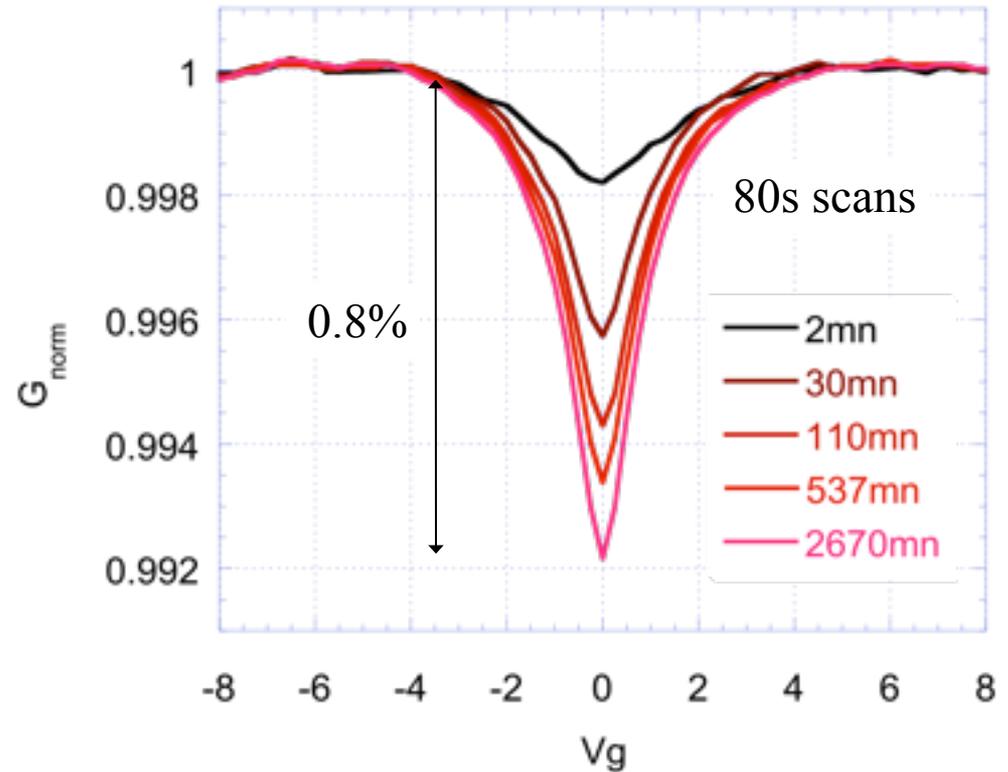
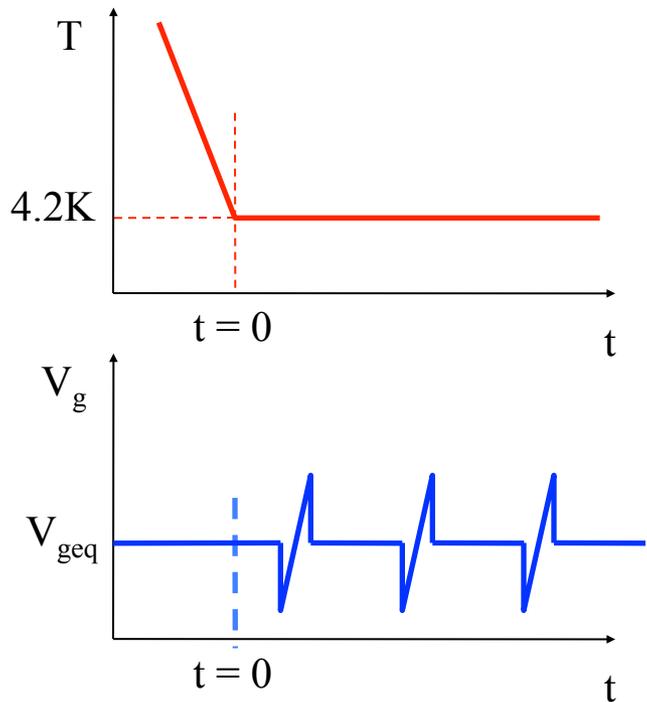
Never ending slow conductance relaxation after a quench



# Out of equilibrium effects: field effect anomaly

$G(t, V_g)$  after a quench at 4.2K

$R_{\square} = 30M\Omega$  at 4.2K

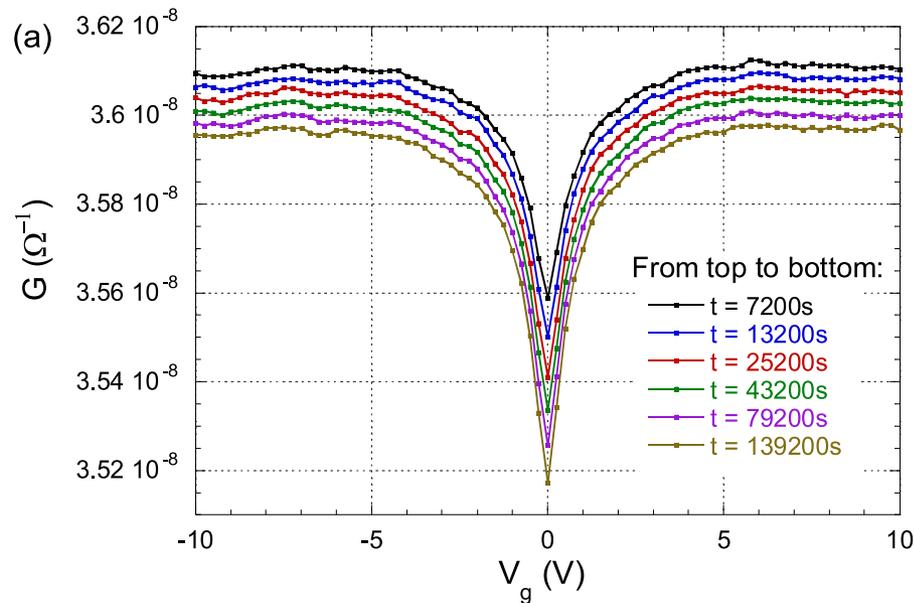


- Field effect anomaly (the “cusp” or “dip”)

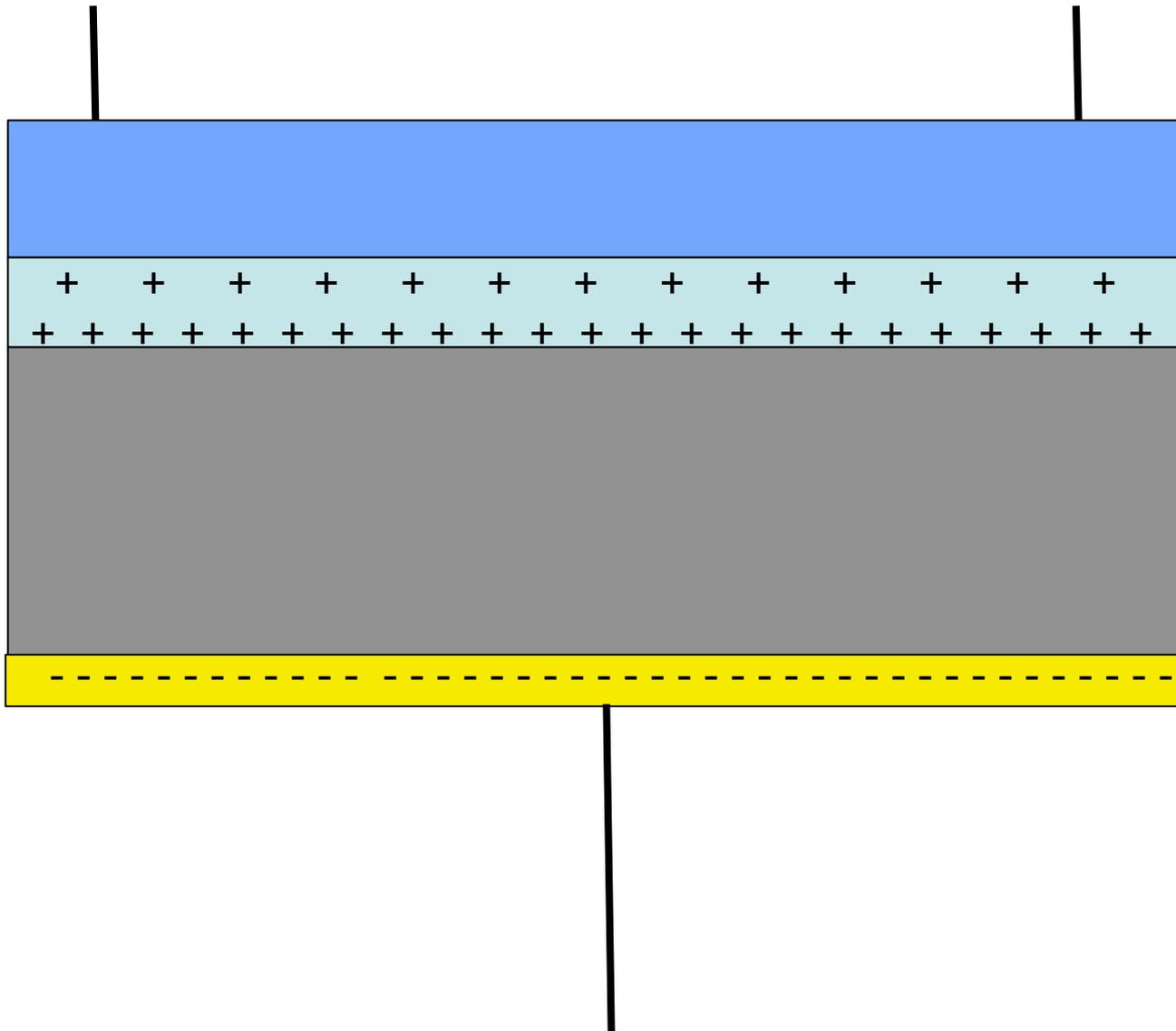
- Amplitude grows like  $\ln(t)$

# Out of equilibrium effects: thickness dependence

Baseline relaxation in «thick enough» samples:



- signature of a finite screening length ( $L_{\text{screen}}$  around 10 nm)

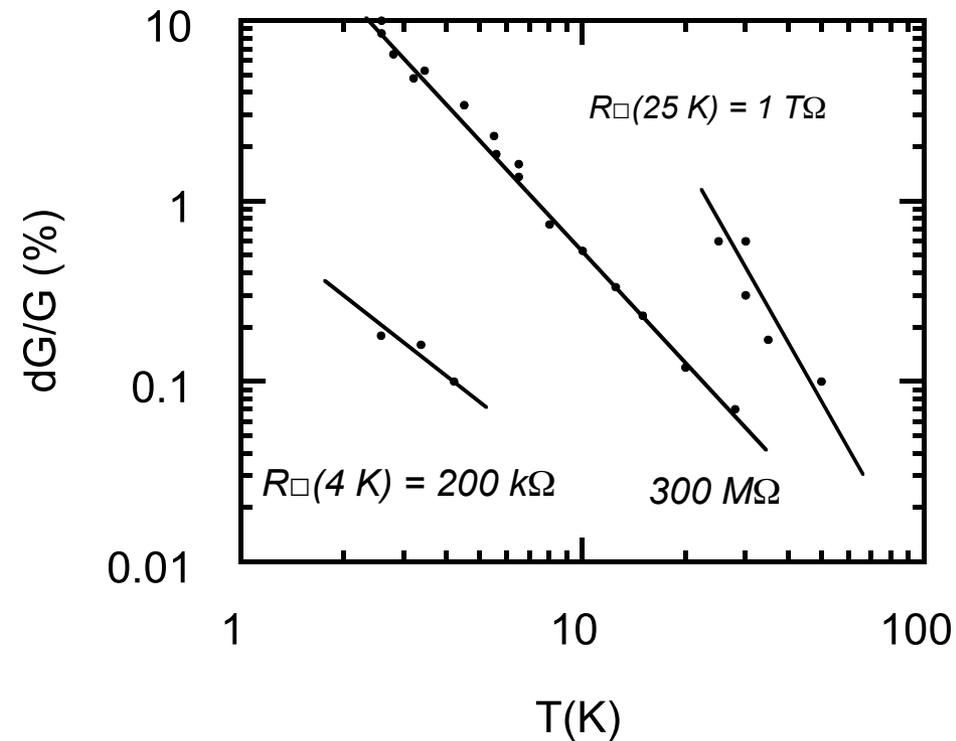


**Granular  
Aluminium**

**Gate**

# When do we see this anomaly ?

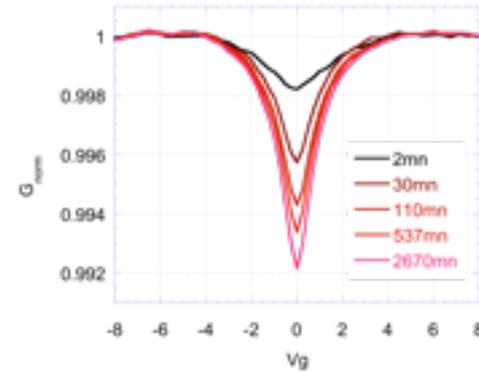
- the anomaly is **always** seen in insulating films
- it is most prominent (in %):
  - at low T (most measurements at 4K)
  - in more insulating samples



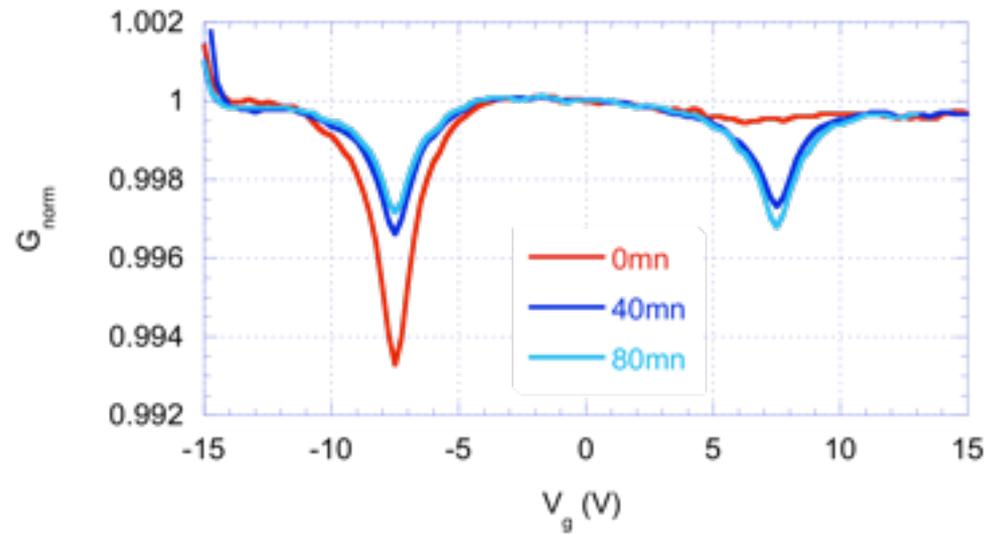
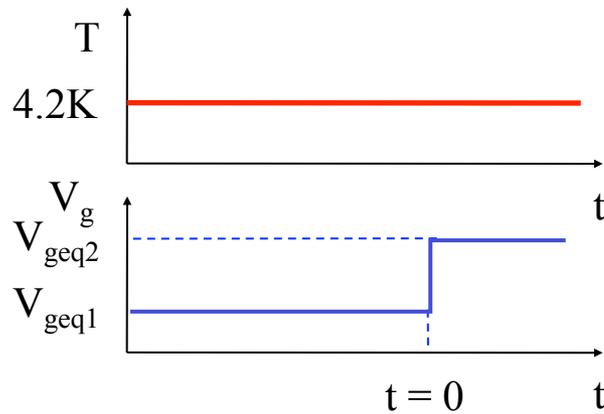
- for practical reasons we study samples where the anomaly is not so large ( $\leq 1\%$ ) but it can be a large effect (more than 10%)

# Cusp dynamics

Recall: after a cooling



After a gate voltage change:

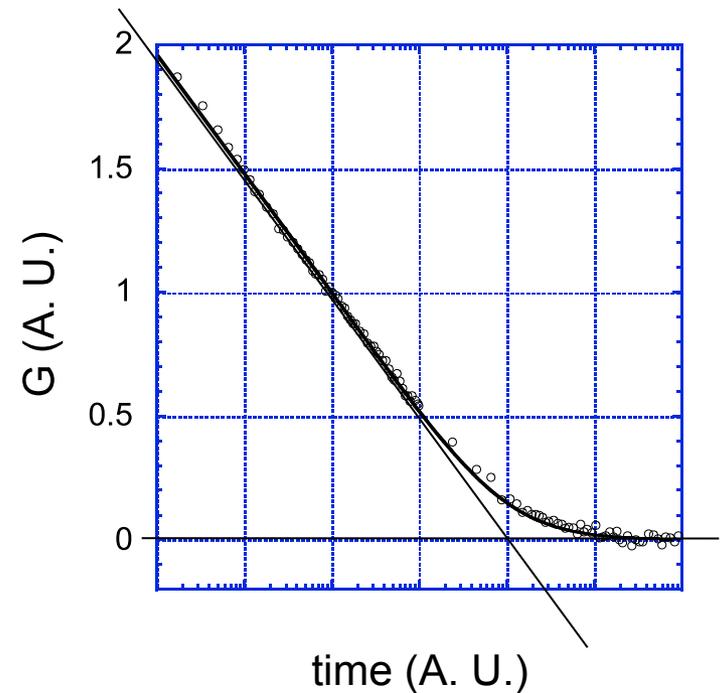
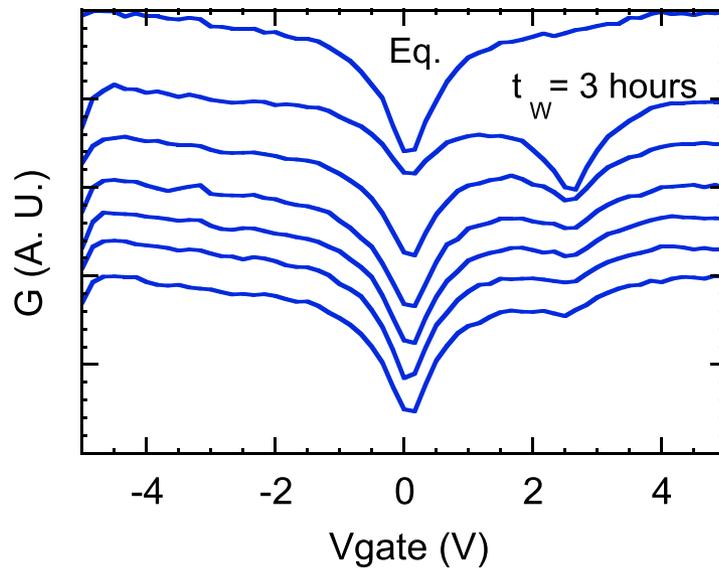


# Is the dynamics activated ?

Is the dynamics accelerated when  $T$  is increased ? (it would explain why the dip becomes very faint)

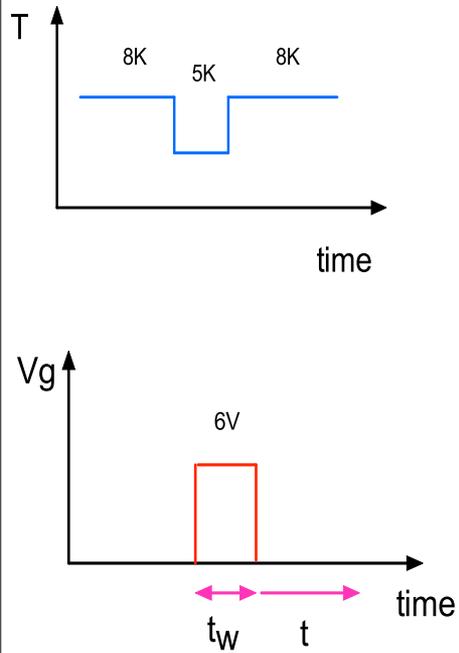
But how to detect a change of the dynamics if it has no characteristic time ?

→ look at the **erasure time** of a previously formed dip

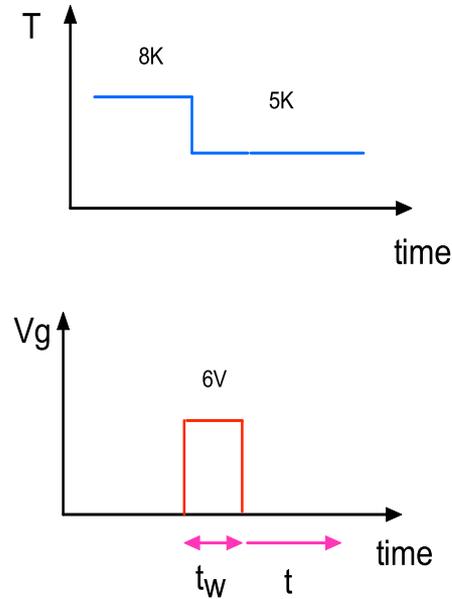


# Is the dynamics activated ?

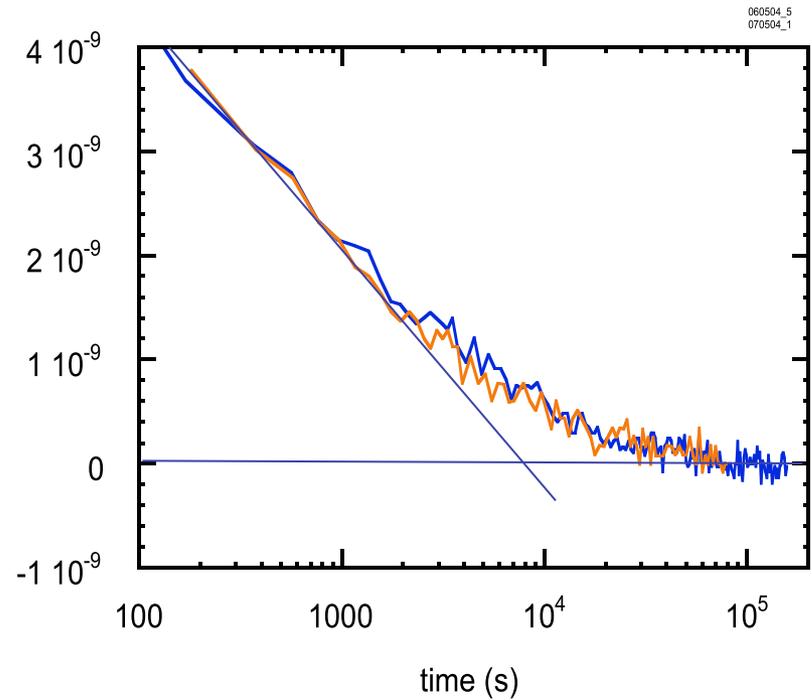
I



II



G



⇒ dynamics is not activated

## OUTLINE

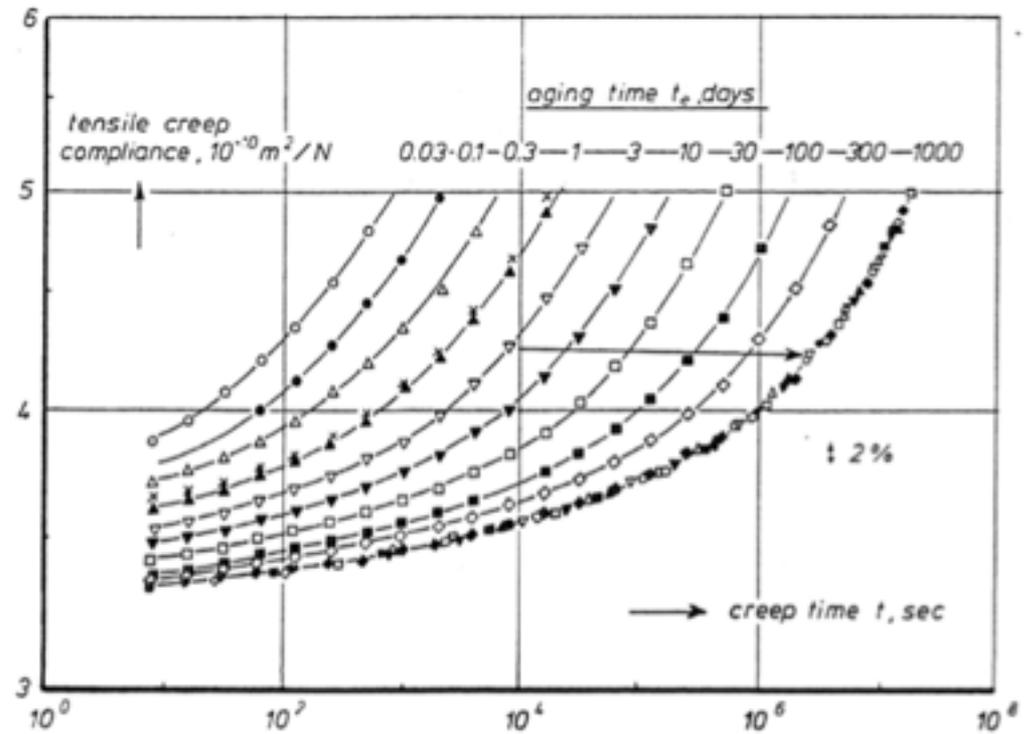
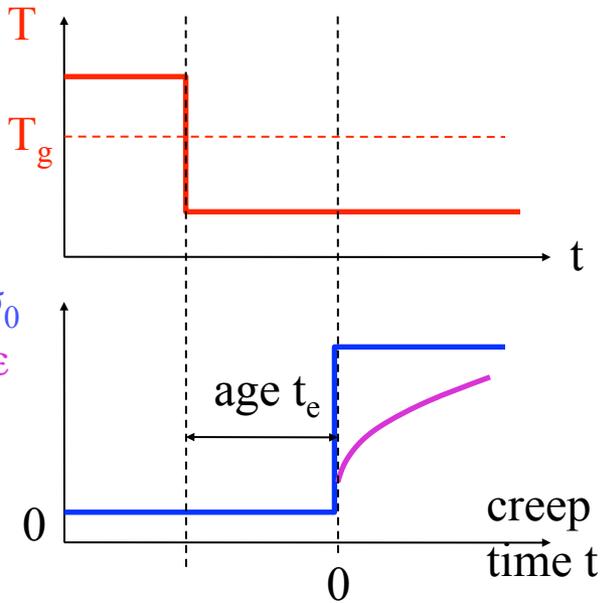
- how it started
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# If it is a glass ... does it age ?

## AGEING:

Ex: creep tests on polymers

Creep compliance  $(t) = \epsilon(t) / \sigma_0$

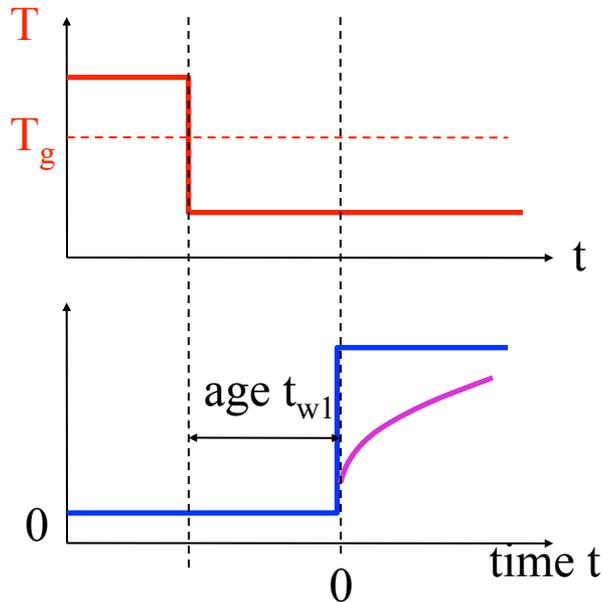


The dynamics depends on time: the « older » the system, the slower the response to a stimulus !

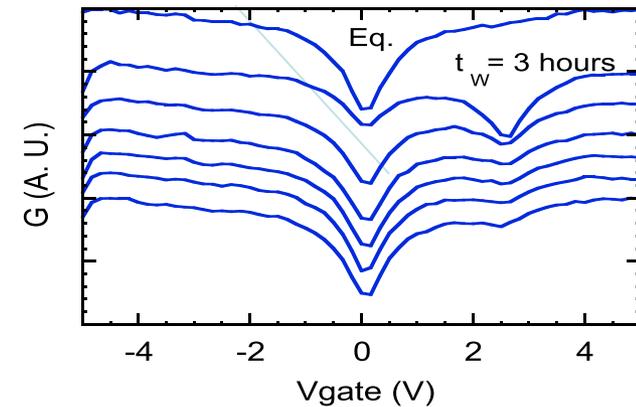
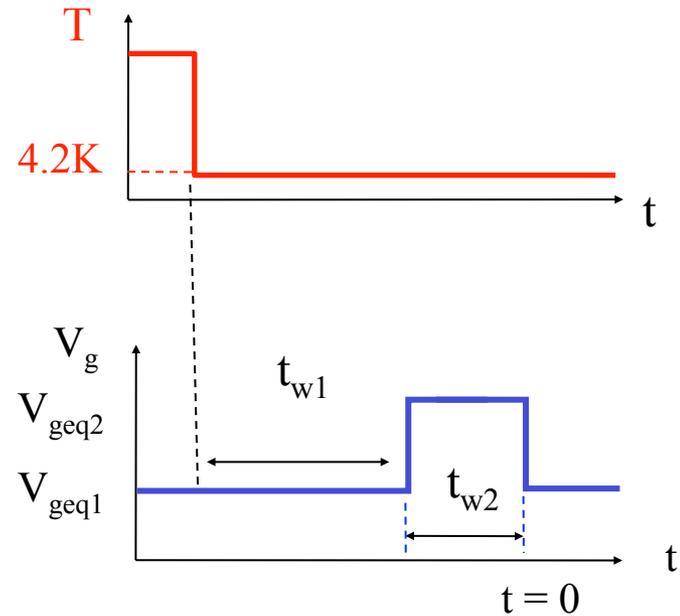
*PVC quenched from 90°C to 40°C (T<sub>g</sub>=80°C)  
L.C.E Struik, 1978*

# « ageing » and « Two dip » protocols

## Standard « ageing » protocol

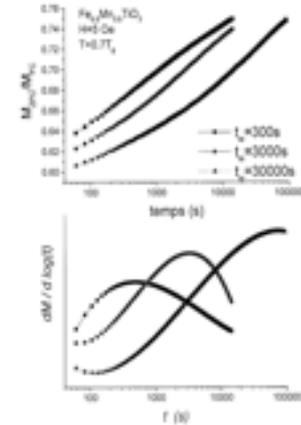
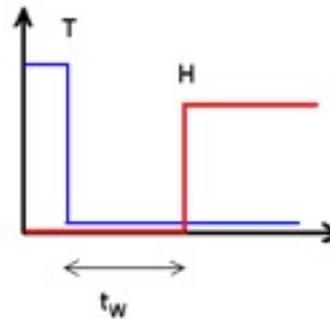


## « Two dips » protocol

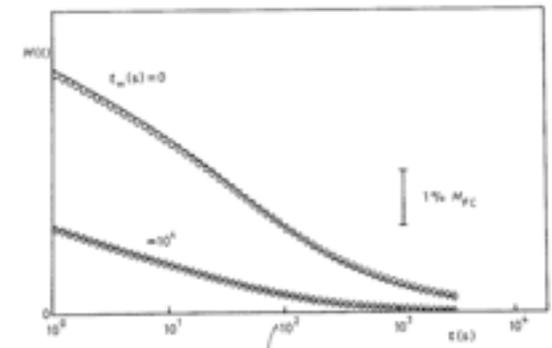
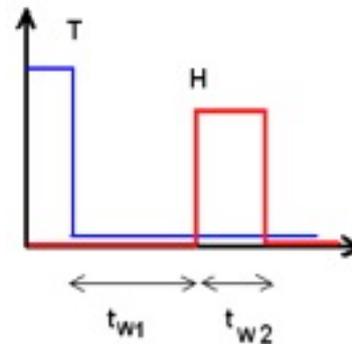


# equivalent spin glasses protocols

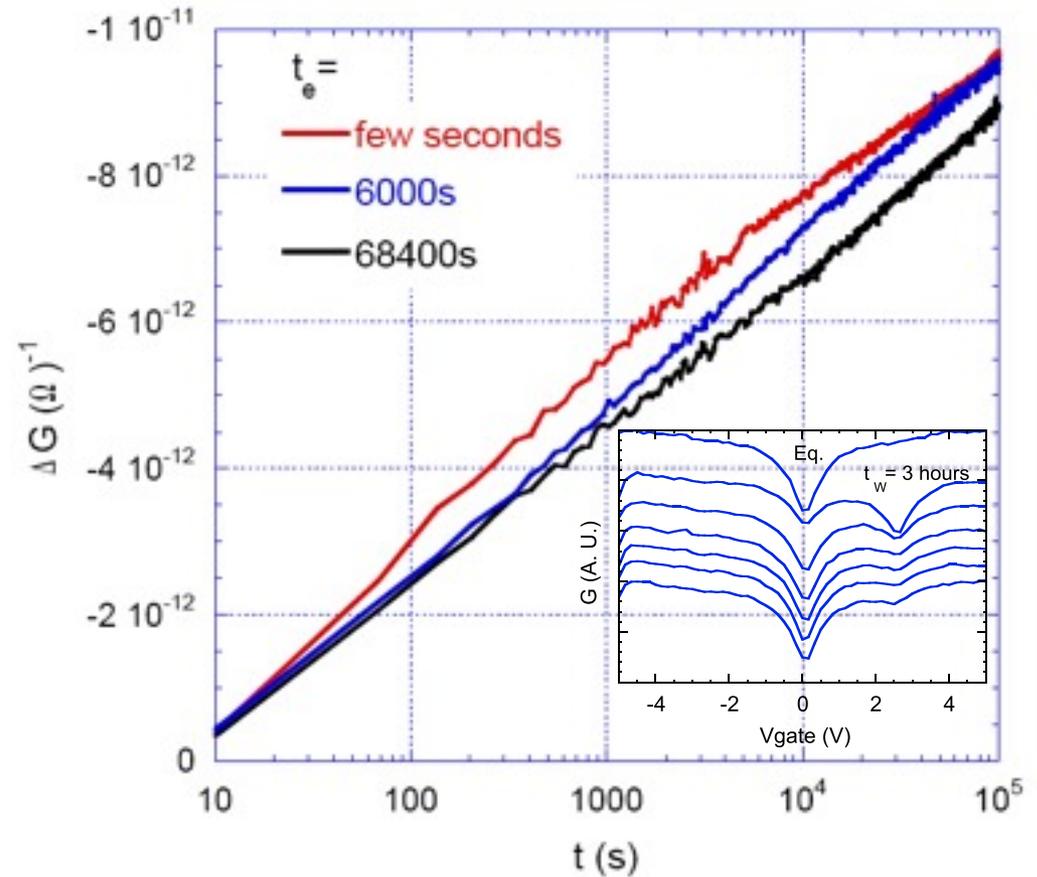
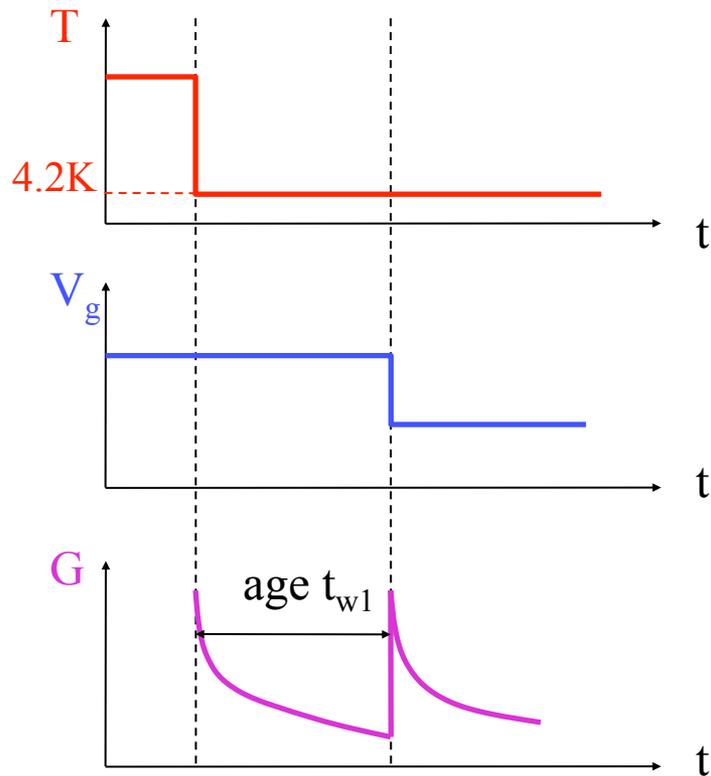
\* Zero field cooled relaxation (ZFC):



\* Isothermal remanent magnetization (IRM):



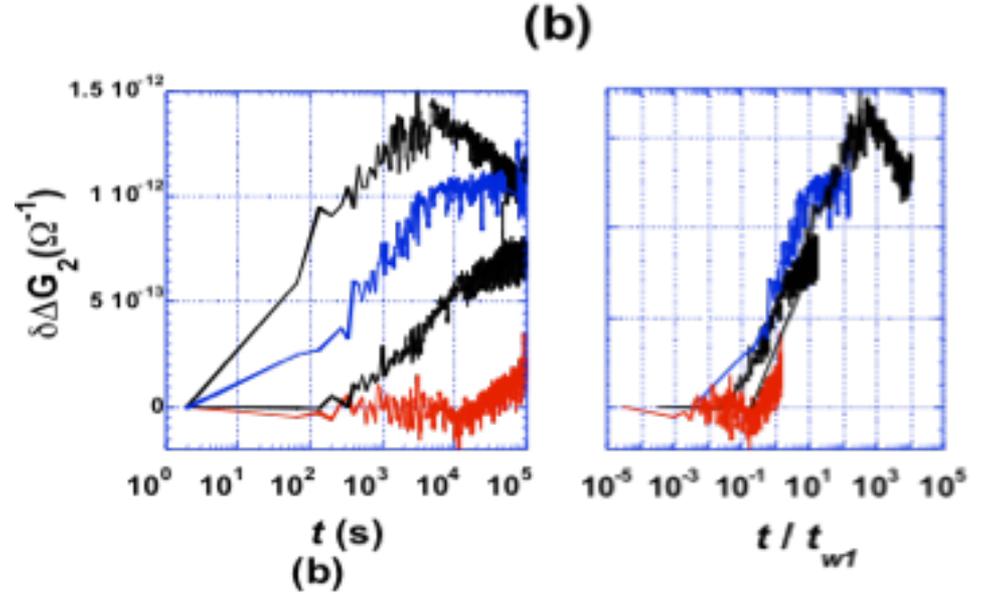
# Standard ageing protocol (1)



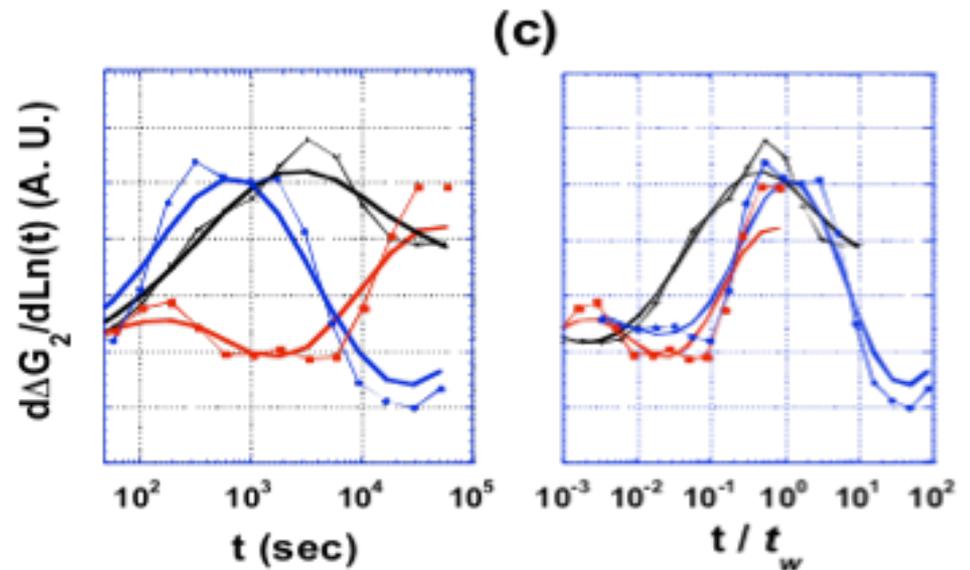
New dip growth: **NOT** like  $\text{Ln}(t)$

# Standard ageing protocol (2)

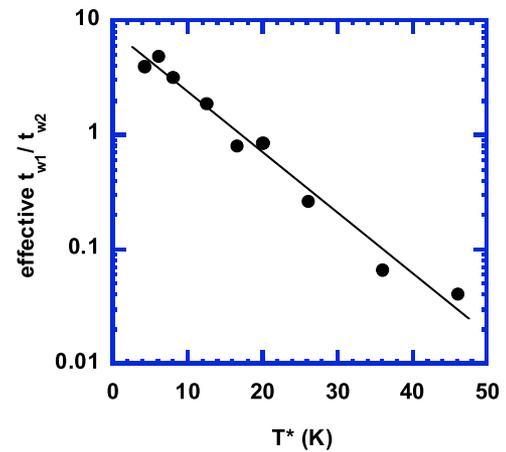
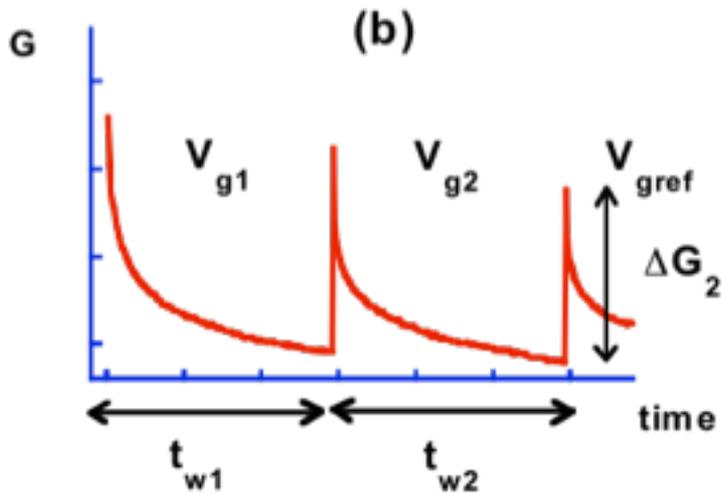
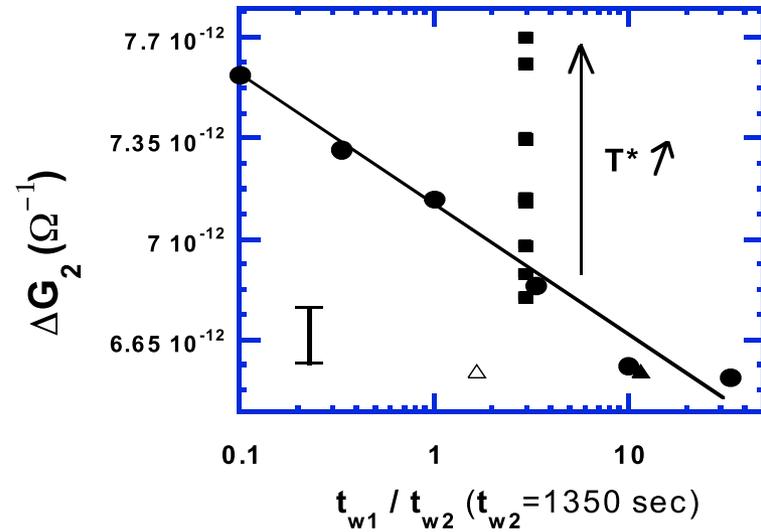
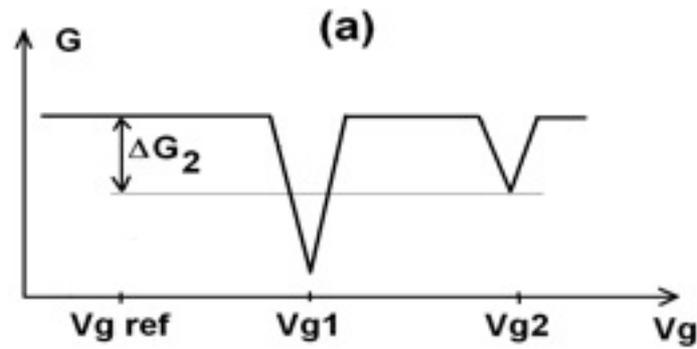
Departures from pure  $\text{Ln}(t)$  relaxation scale with  $t_{w1}$



Effective relaxation time distribution  $d\Delta G_2/d\text{Ln}(t)$  scales with  $t_{w1}$

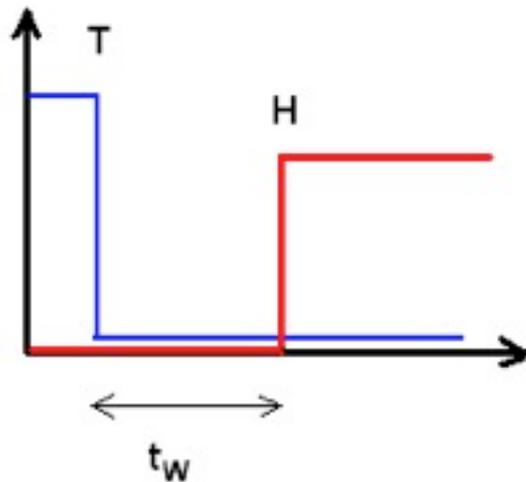


# Rejuvenation by annealing

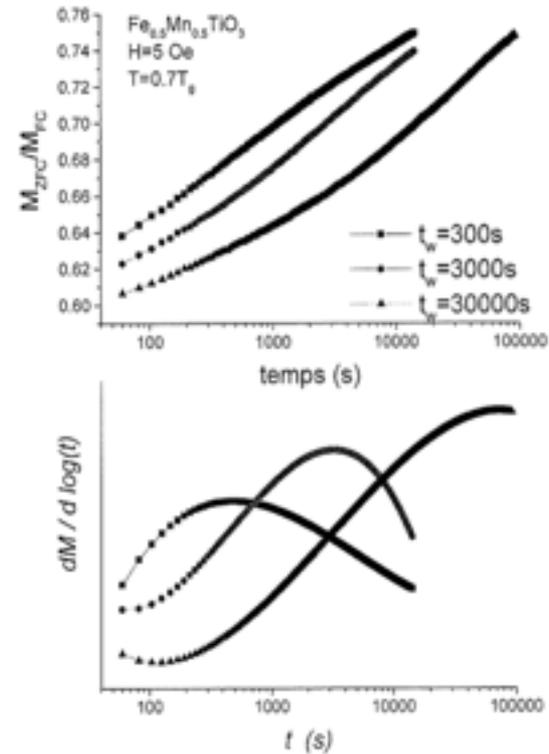


# Example of spin glasses

## Zero field cooling relaxation



## Magnetization (t)

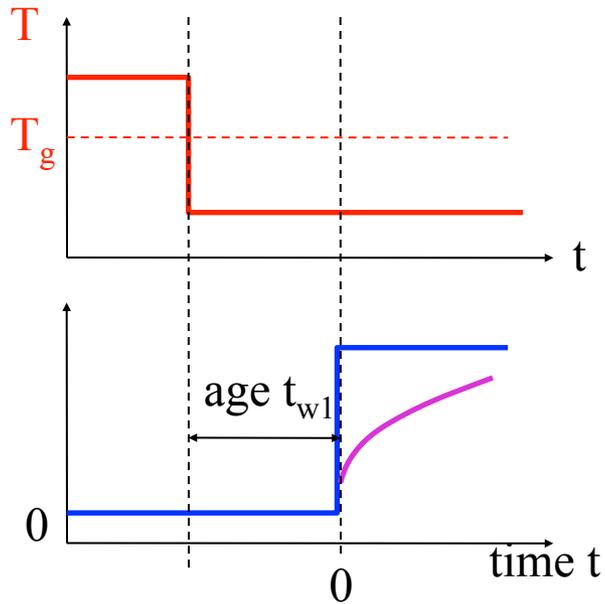


*E. Vincent, 2006*

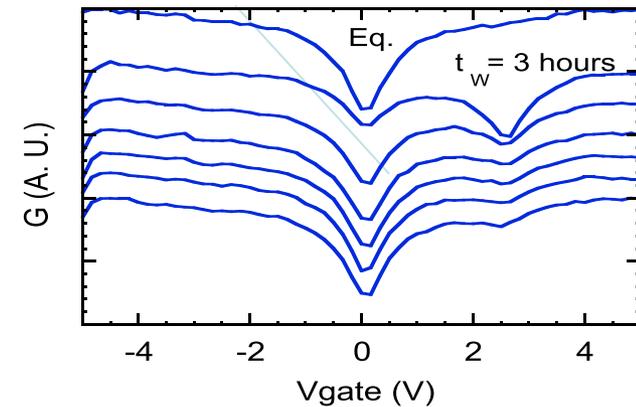
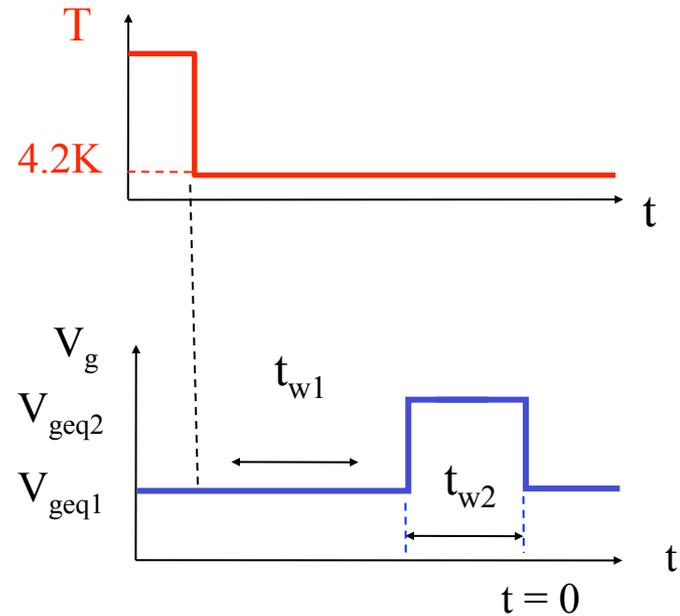
The age of the system is printed in its relaxation time distribution

# « ageing » and « Two dip » protocols

## Standard « ageing » protocol

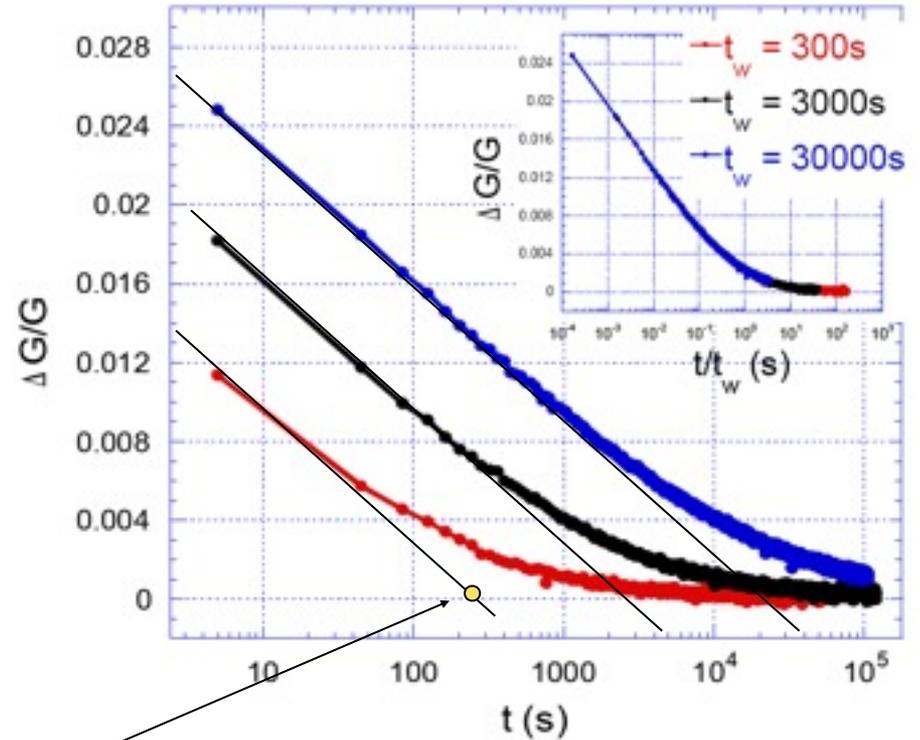
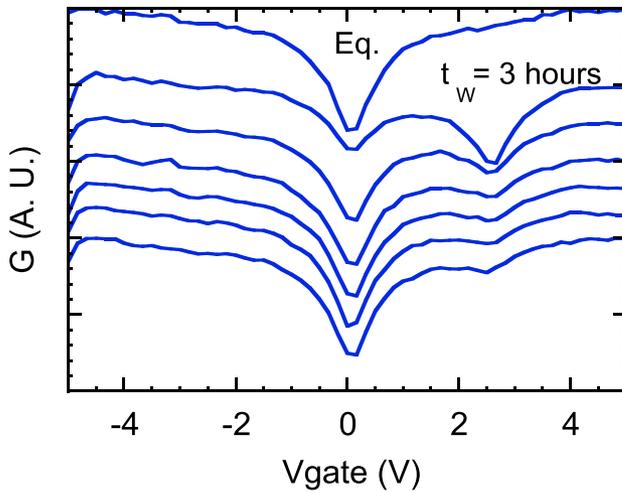


## « Two dips » protocol



# Two dip protocol: very «old» system

« Two dips » protocol

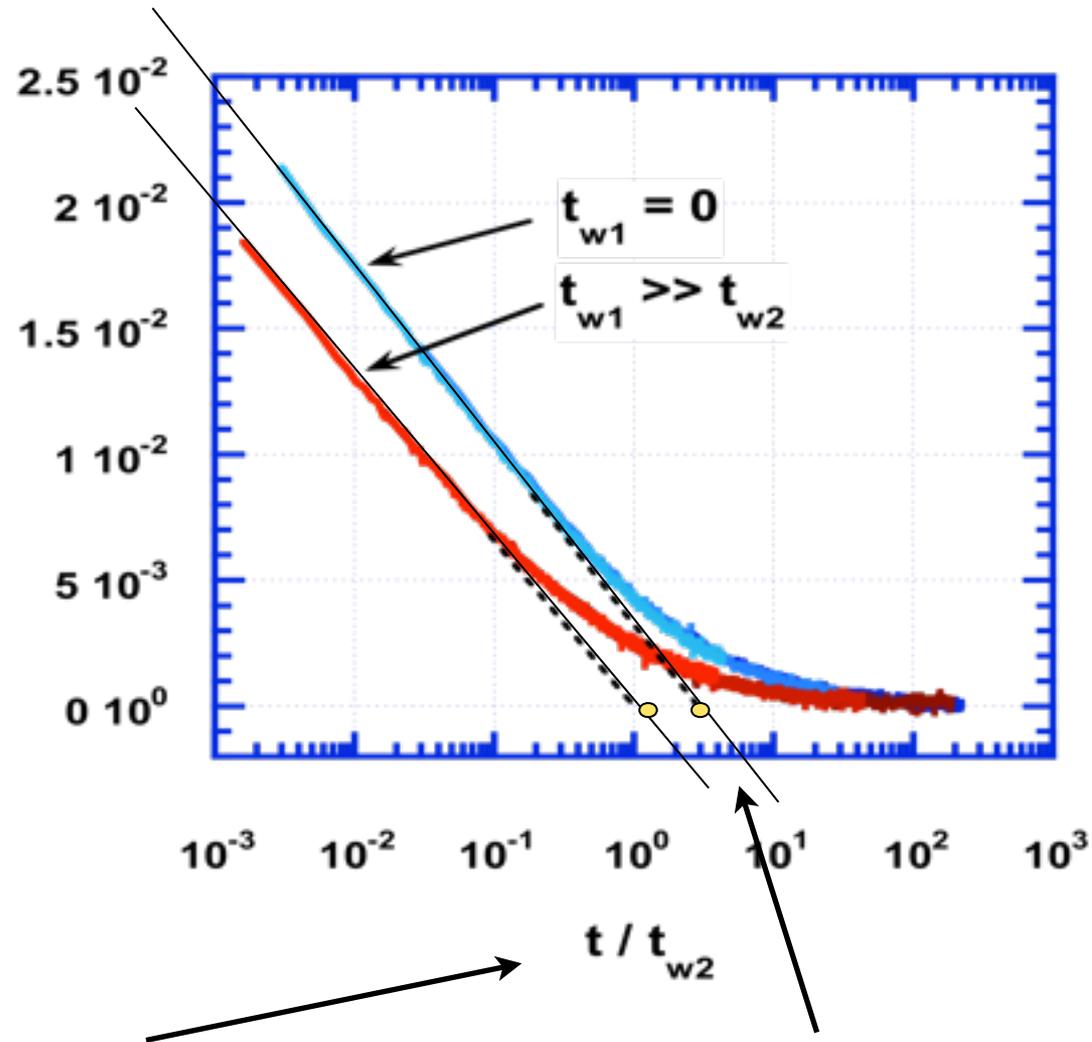
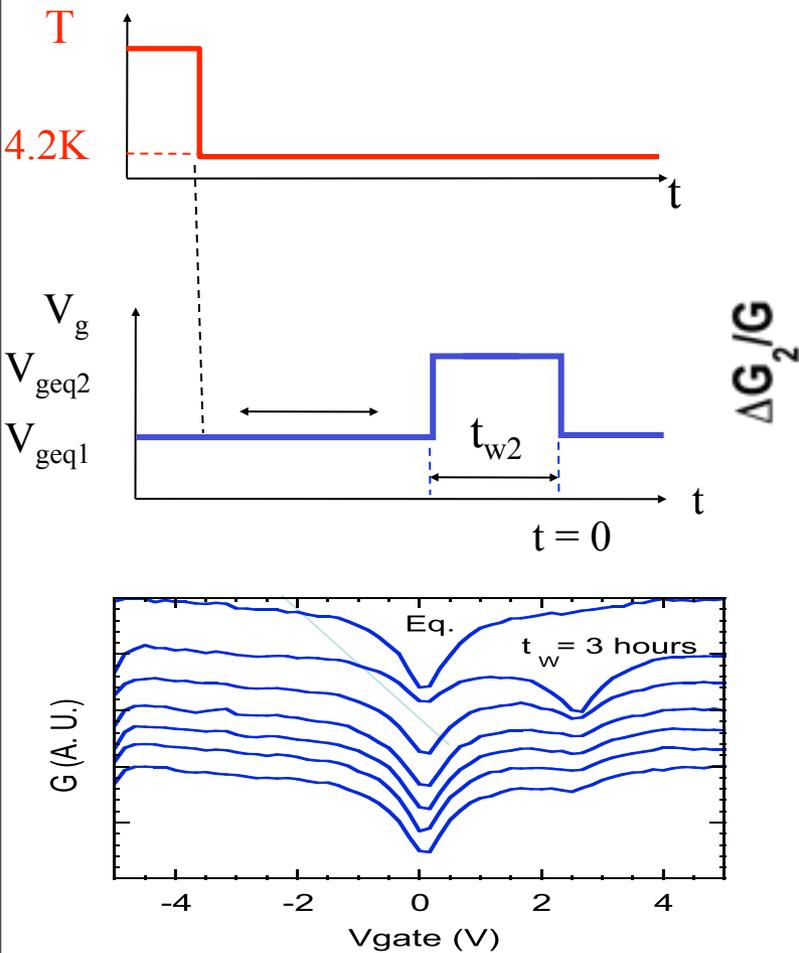


Erasure time

$t/t_{w2}$  scaling

# Two dip protocol: «young» (ageing) system

## « Two dips » protocol



$t/t_{w2}$  scaling:  
memory of  $t_{w2}$

value of  $t/t_{w2}$  erasure time:  
memory of  $t_{w1}$

# A simple quantitative approach when $t_{w1} \gg t_{w2}$

A simple model can reproduce the data:

- collection of independent reversible « degrees of freedom »
- additive effect on G
- tunnel  $\rightarrow \ln(\tau_i)$  has a broad (flat) distribution

Then:

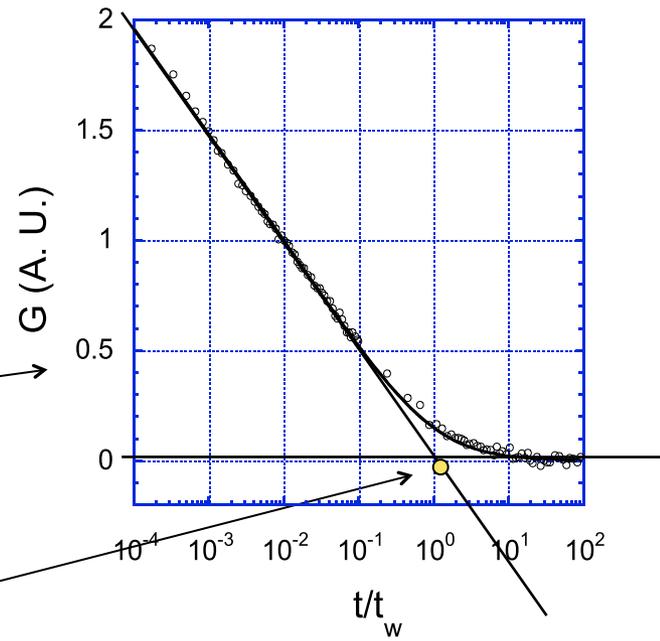
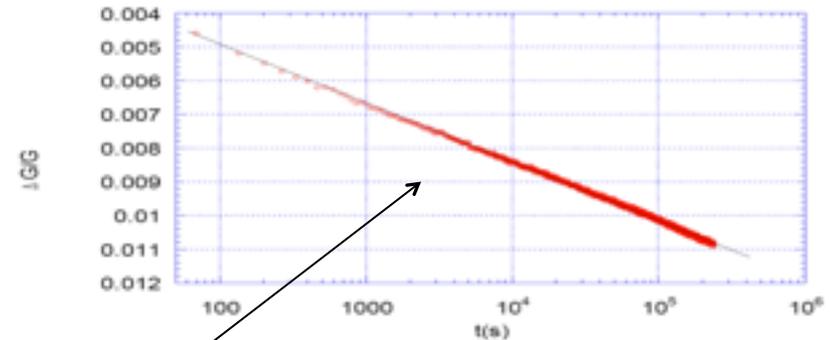
*writing:*

$$\Delta G(t < t_w) = -\Delta G_0 \sum_i (1 - \exp(-\frac{t}{\tau_i}))$$

*erasure:*

$$\Delta G(t > t_w) = \Delta G_0 \sum_i (1 - \exp(-\frac{t_w}{\tau_i})) \exp(-\frac{t - t_w}{\tau_i})$$

$$\Delta G(t > t_w) = -\Delta G_0 \ln(1 - \frac{t_w}{t})$$



erasure time

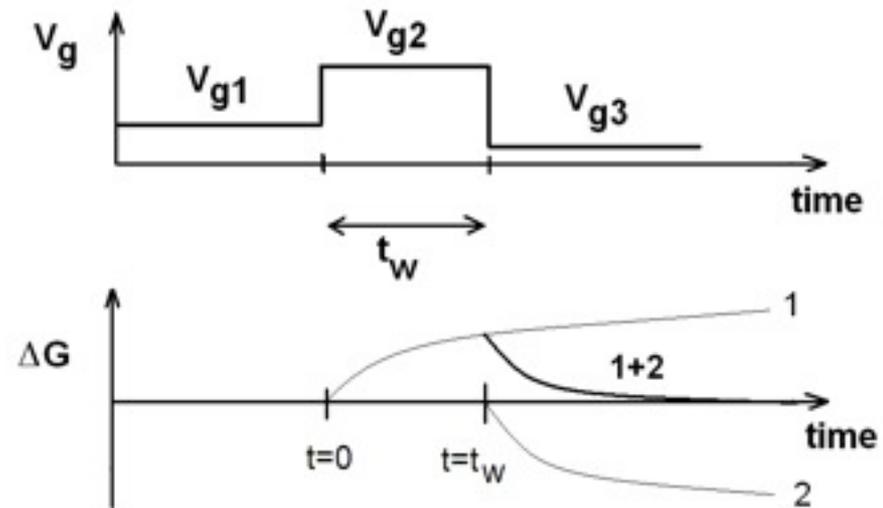
# «Superposition principle»

*erasure:*

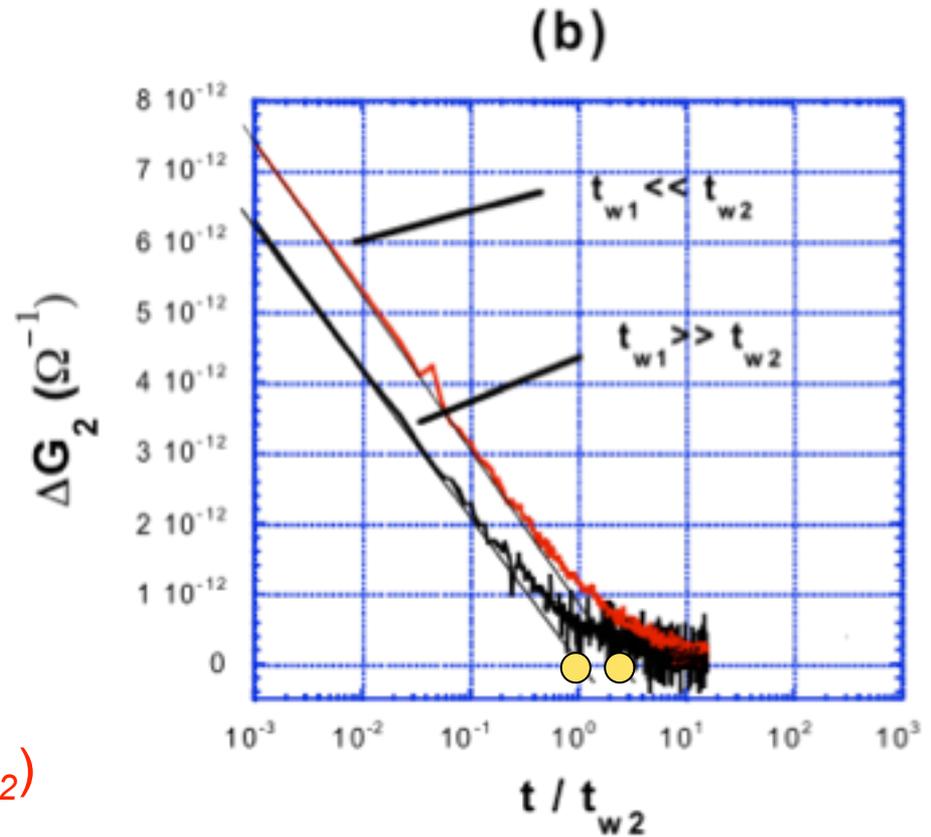
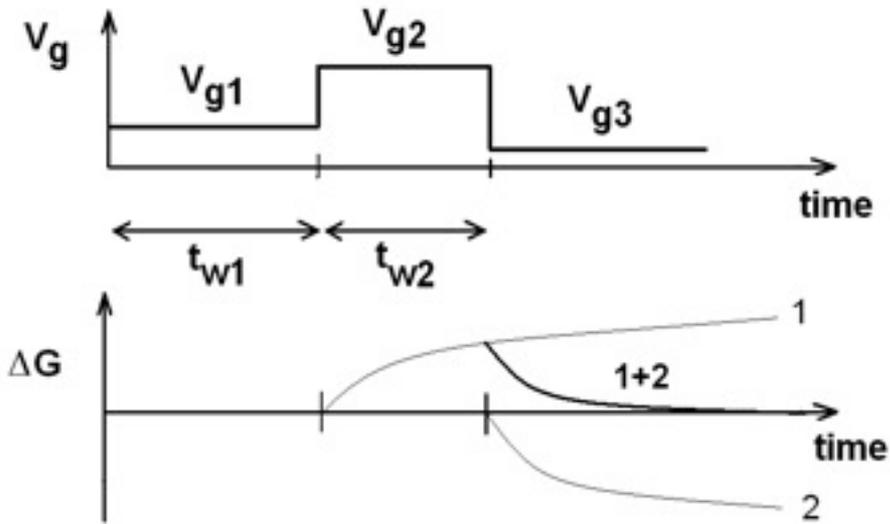
$$\Delta G(t > t_w) = \Delta G_0 \sum_i (1 - \exp(-\frac{t_w}{\tau_i})) \exp(-\frac{t - t_w}{\tau_i})$$

$$\Delta G(t > t_w) = \Delta G_0 \sum_i (1 - \exp(-\frac{t}{\tau_i})) - (1 - \exp(-\frac{t - t_w}{\tau_i}))$$

$$\Delta G(t > t_w) = \Delta G_0 (Ln(t) - Ln(t - t_w))$$



# «Superposition principle»



Application of the principle for  $G(V_g = V_{g2})$

# A simple quantitative approach of ageing

- collection of independant reversible « degrees of freedom »

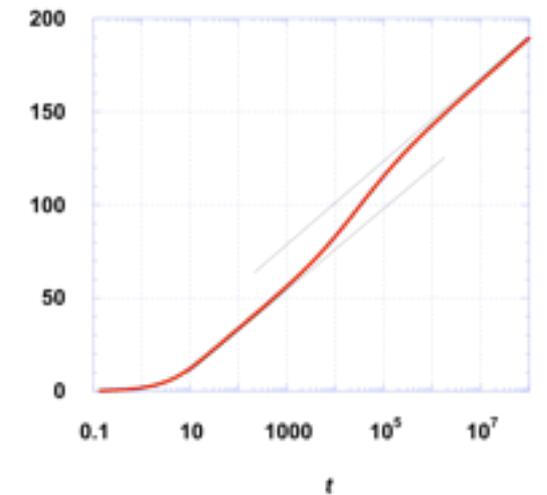
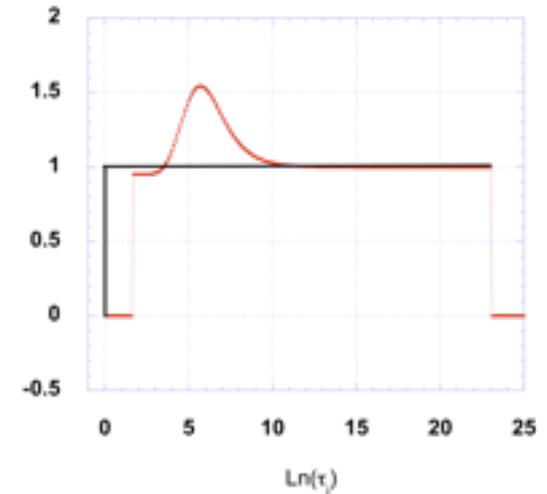
- additive effect on G

-  $\text{Ln}(\tau_i)$  has a broad (flat) distribution

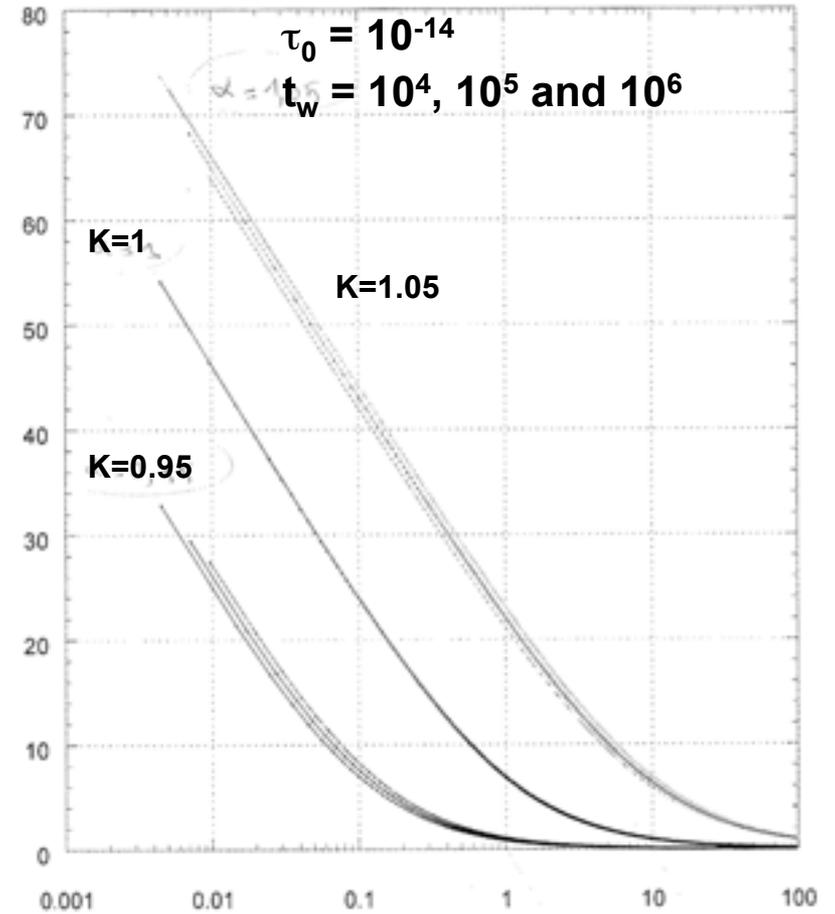
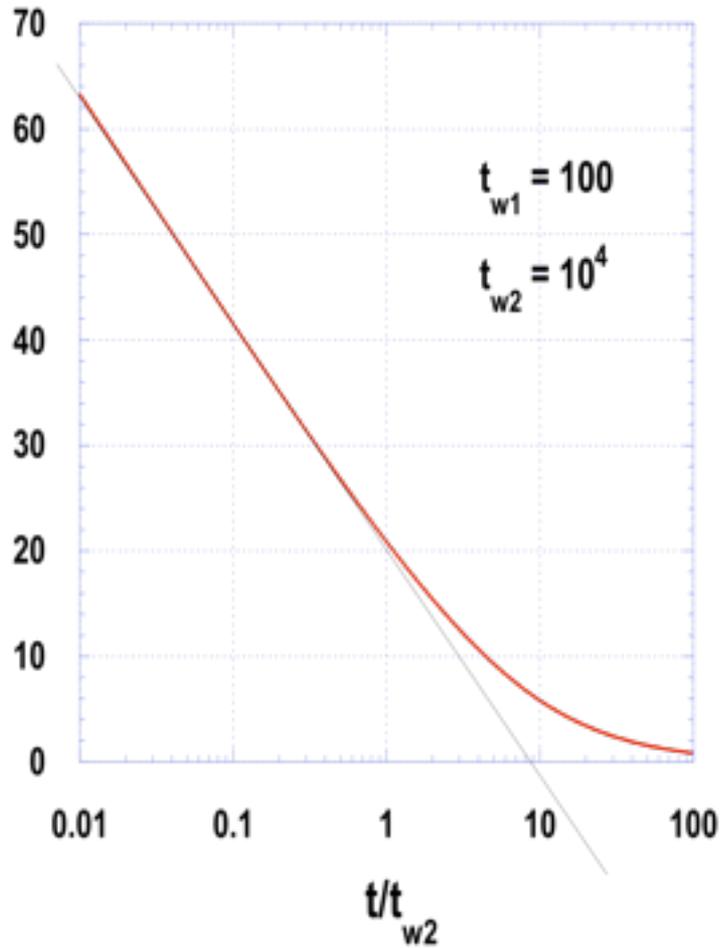
-  $\tau_{i \rightarrow} \neq \tau_{i \leftarrow}$

Suppose:

$$\tau_{i \leftarrow} = \tau_0 \exp(\xi_i) \text{ and } \tau_{i \rightarrow} = \tau_0 \exp(k * \xi_i)$$



# A simple quantitative approach of ageing



# OUTLINE

- how it started
- some aspects of the « glassiness » in granular Al
- is ageing present ? (is it a glass?)
- questions

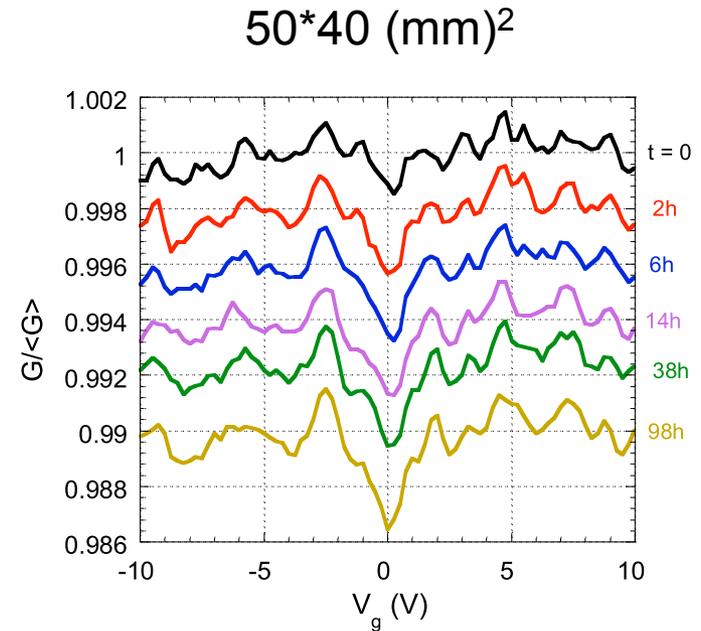
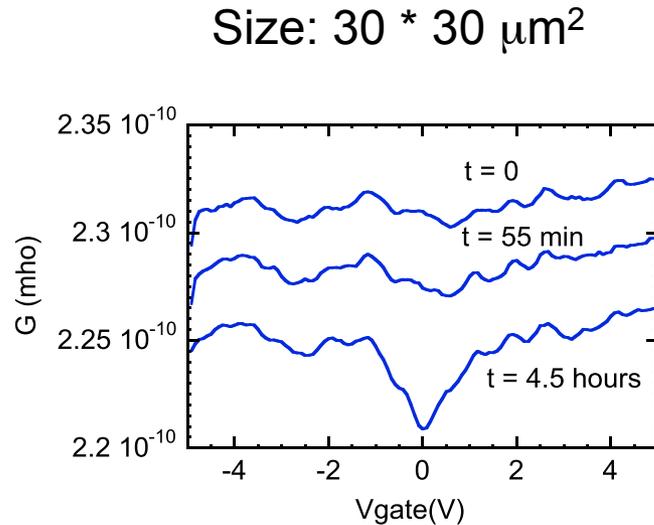
# Is this glass purely electronic ?

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- competing « extrinsic » (non electronic) scenarios can be envisaged (slow atomic or ionic processes influencing the conductance)
- are there indications in favor of the coulomb electron glass ?
- 1) in Indium oxide: **effect of carrier concentration** (varied by changing oxygen concentration)
  - \* systematic effect of carrier concentration on the field effect anomaly width
  - \* the dynamics is also influenced by the carrier concentration (fast erasure of a formed field effect anomaly by a high enough  $V_g$  change)

# Is this glass purely electronic ?

- 2) slow relaxation in mesoscopic samples :



- mesoscopic fluctuations (fluctuations of percolation path as a function of  $V_g$ ) and the cusp coexist

- both seem to have **very different time scales** (disorder seems totally frozen)  $\rightarrow$  **may be consistent with electron glass** (cusp slow relaxation not due to disorder (atoms) relaxation)

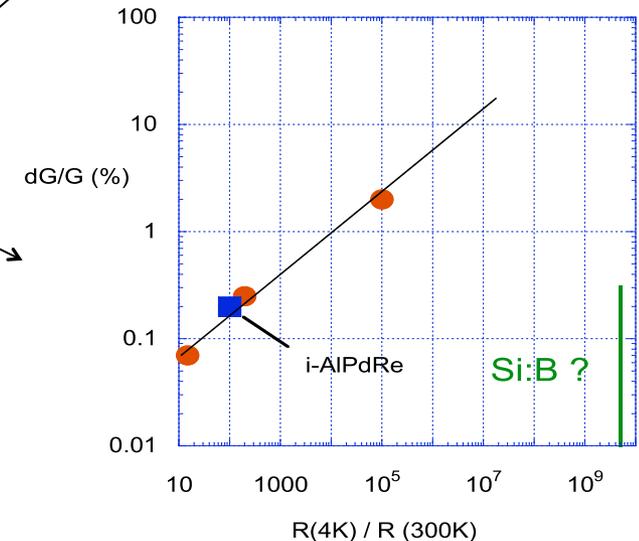
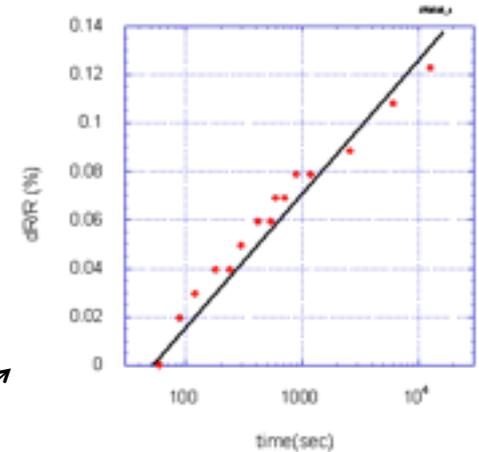
# Is this glass purely electronic ?

- 3) systematics in other materials:

Up to now:

- studied in: indium oxide, granular aluminium
- seen in: granular gold, ultra thin Pb on a-Ge
- being studied in Ni films (without oxide, strong effect of magnetic field on the dynamics, Aviad Frydman)
- maybe present in icosahedral insulating quasicrystal i-AlPdRe

What do these materials have in common ?



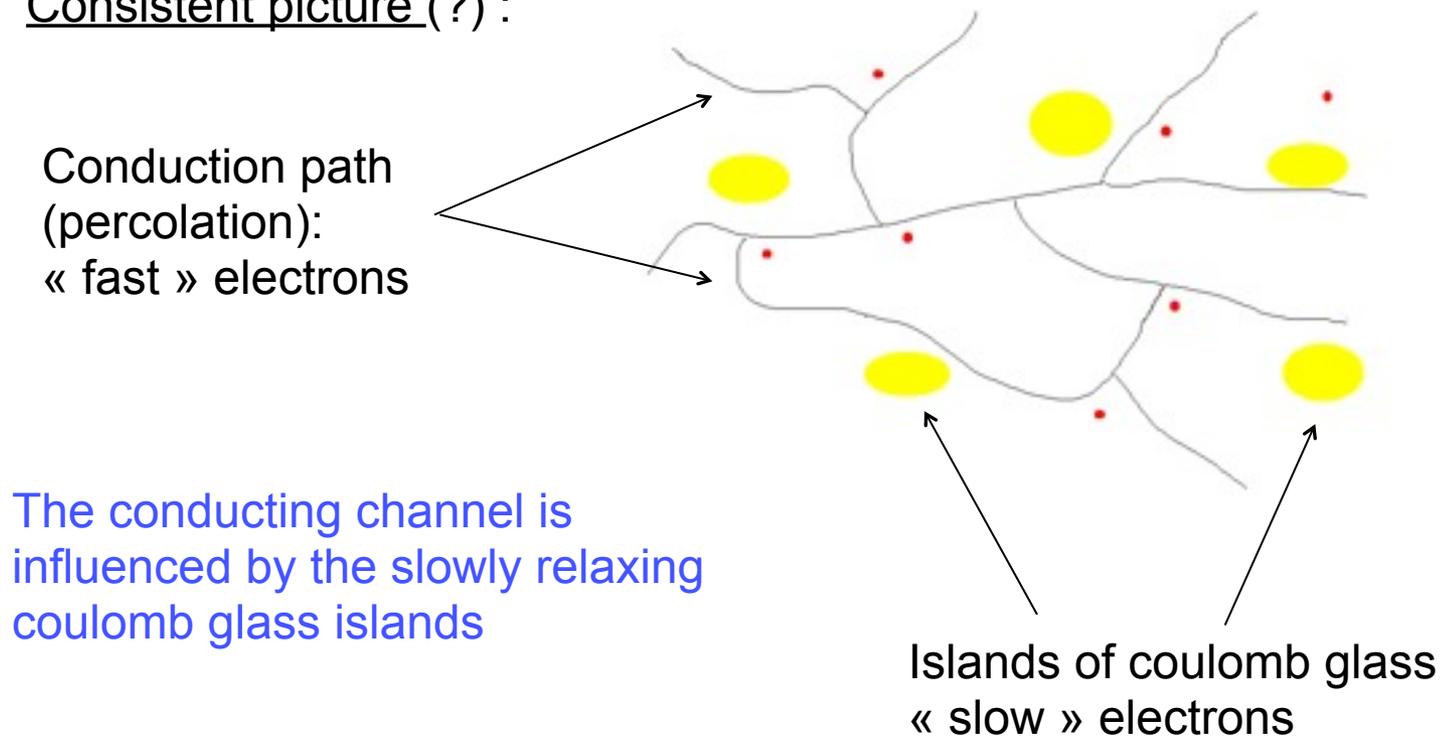
# Only expected in insulating samples ?

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## Apparent paradoxes:

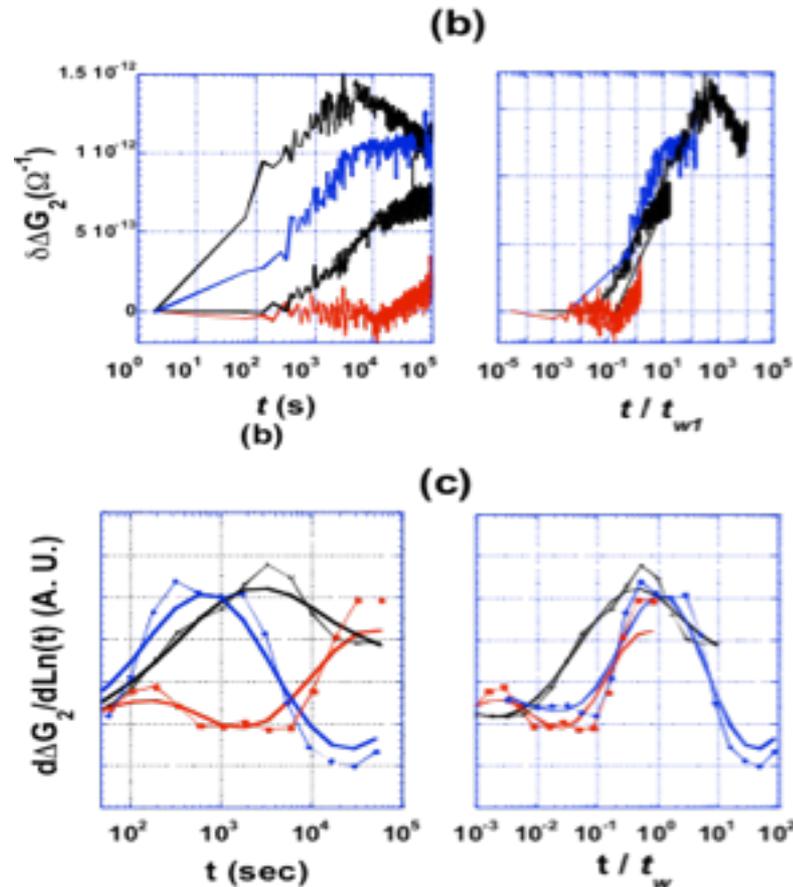
- thermal memory of cusp but not of « back-ground » conductance
- very slow relaxations even for weakly (and metallic?) insulating samples

## Consistent picture (?) :



# Understanding the dynamics ?

- mechanism / significance of the ageing behaviour ?  
(indication for the importance of correlations ?)



What we need:

theoretical predictions\*

\*that can be tested experimentally