Glassy dynamics in porous Si electrical conductivity

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Charge transport in mesoporous Si

Electrochemical etching of p⁺-type Si (free carriers 10^{19} cm^{-3})

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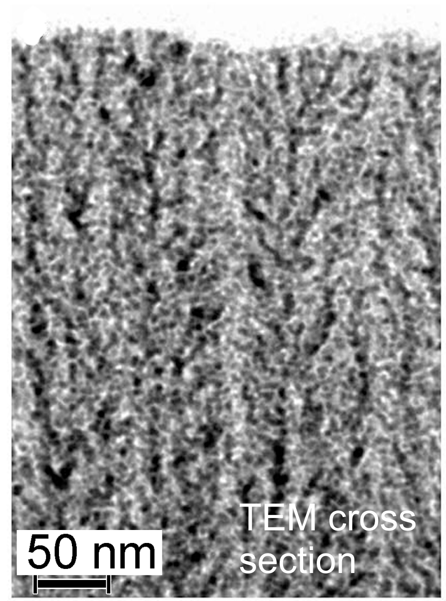
Disordered network of interconnected Si crystallites (typical size 20-100 nm)

Free carriers 10^{18} cm^{-3}, but conductivity drops of 5 orders of magnitude or more



Surface traps (Si dangling bonds); nanoconstrictions

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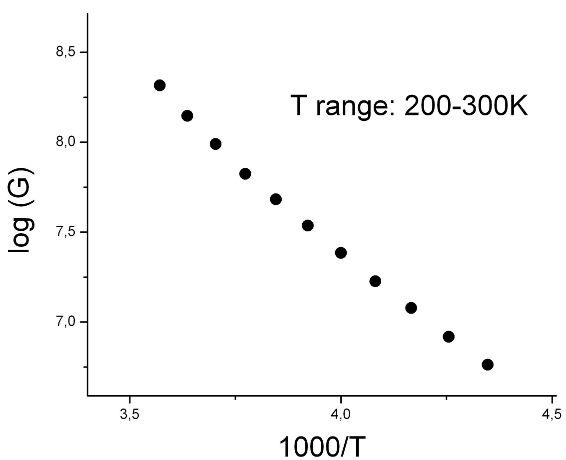
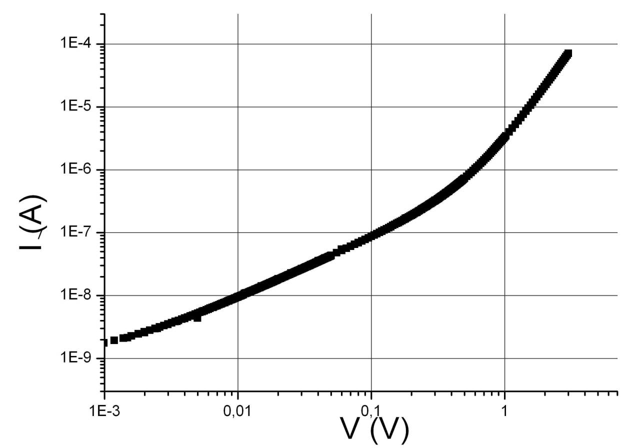
Electrical conductivity limiting mechanism: repulsive charged traps at the nanoconstrictions

[V. Lehmann et al., Thin Solid Films 1995]

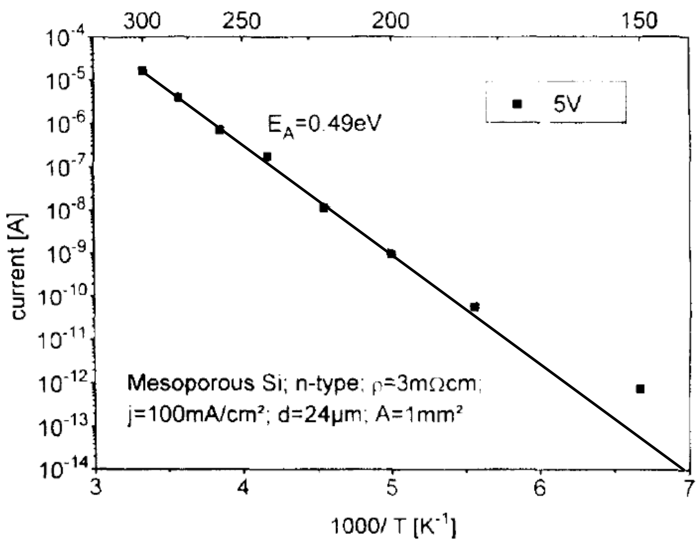


Thermally activated transport: activation energy E_A of the order of hundreds of meV

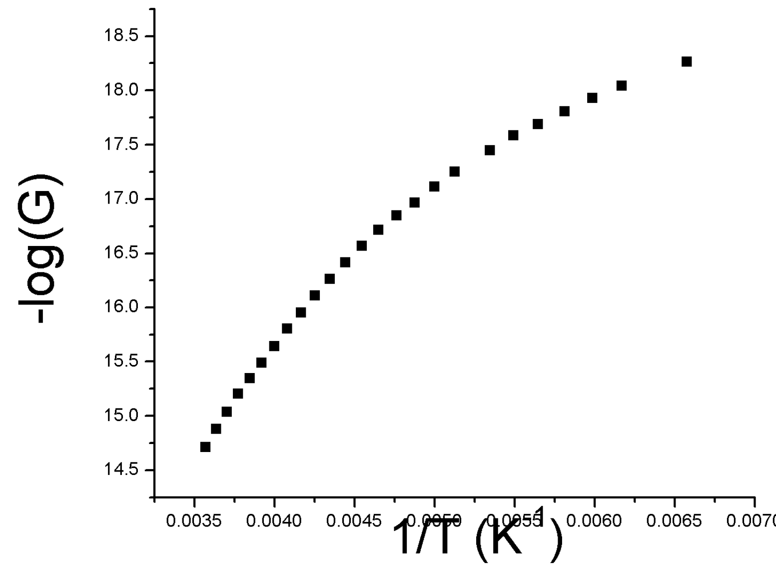


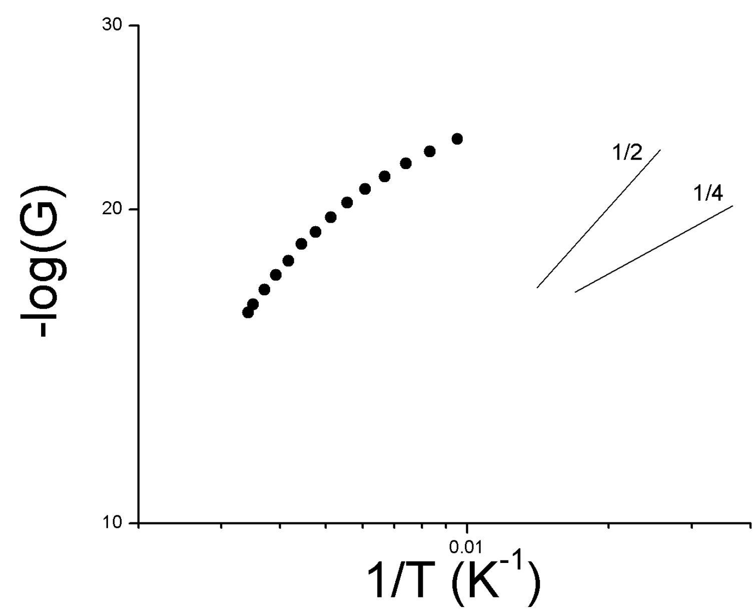
Non-ohmic IV characteristics

Conductance vs T



[V. Lehmann et al., Thin Solid Films 1995]

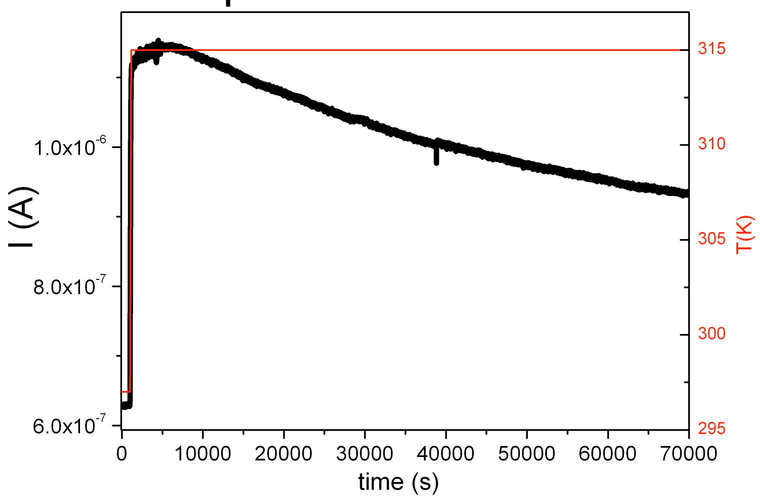




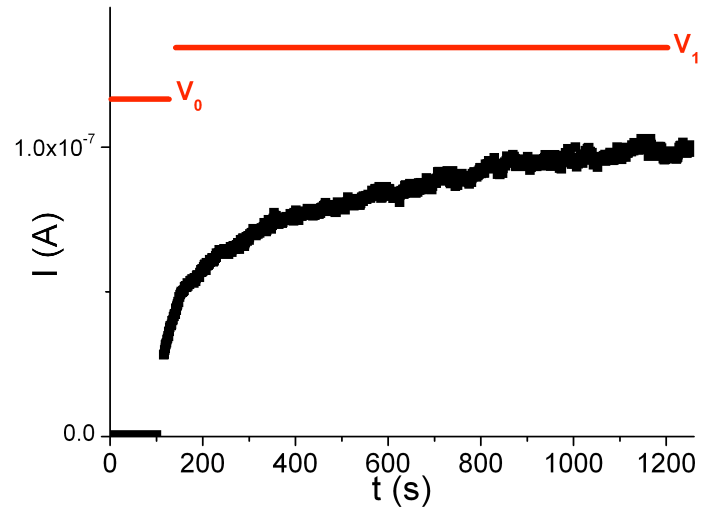
Hopping?

Slow response of electrical conductivity to external *stimuli*

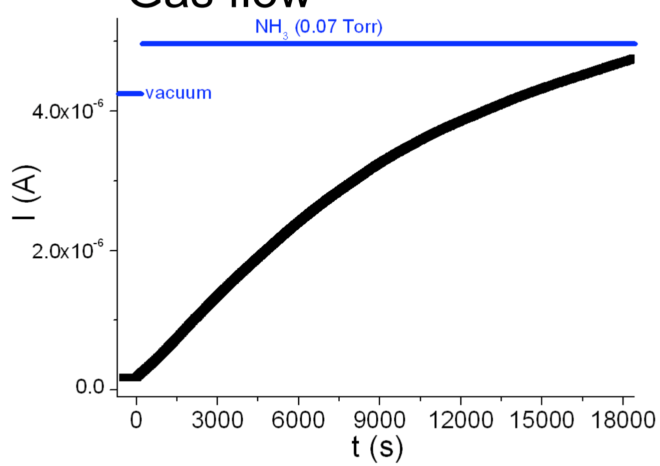
Temperature



Electric field



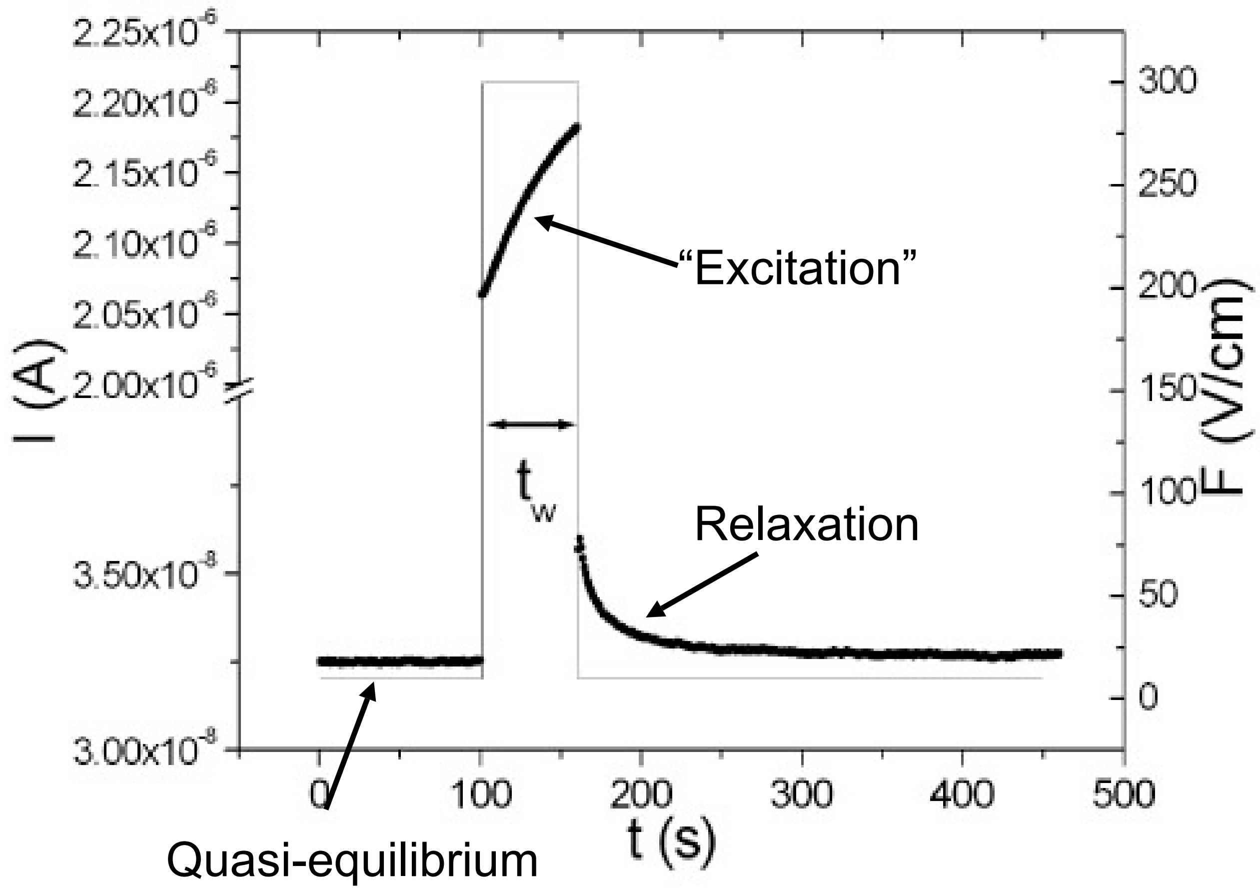
Gas flow



Out of equilibrium system
Non-exponential relaxation
Glassy dynamics?

The "stress aging" protocol

(First proposed for the electron glass in In oxide, Orlyanchik and Ovadyahu, PRL2004)



We use this protocol in order to:

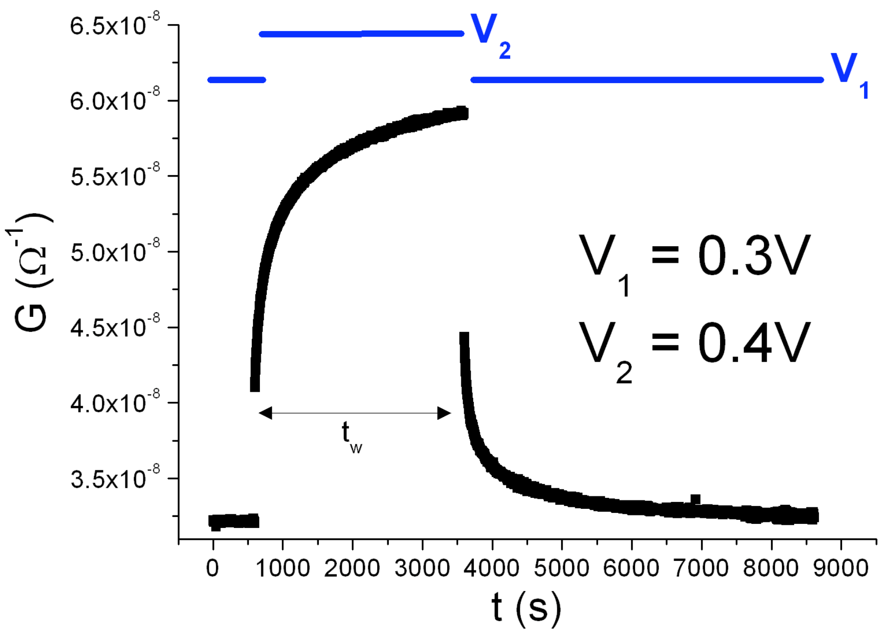
- Study relaxation as a function of t_w

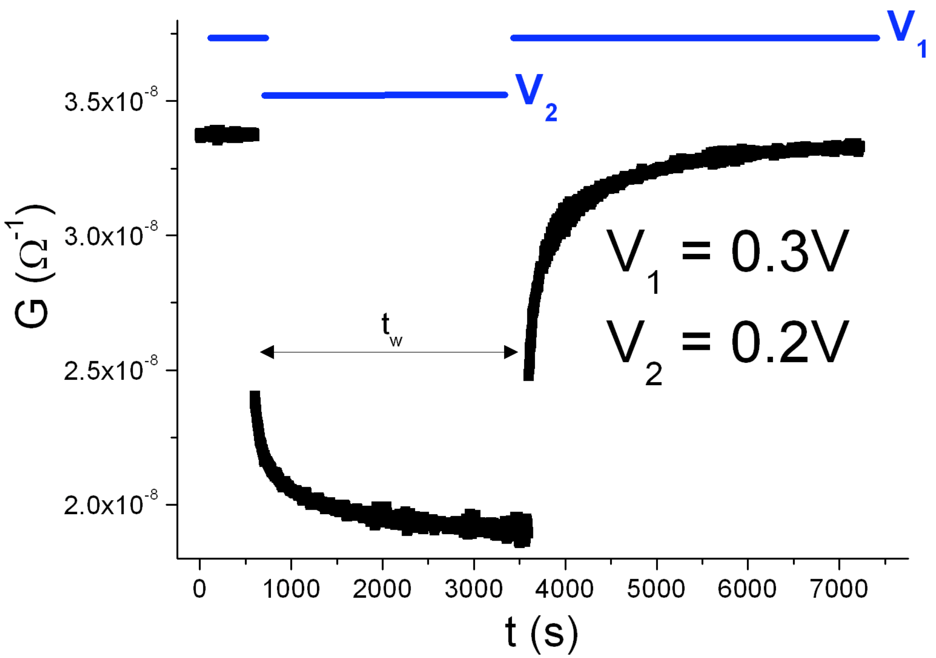
- Find the non-exponential master function describing relaxation

Study relaxation as a function of the temperature

- Draw conclusions on the glassiness of the system

Excitation / relaxation



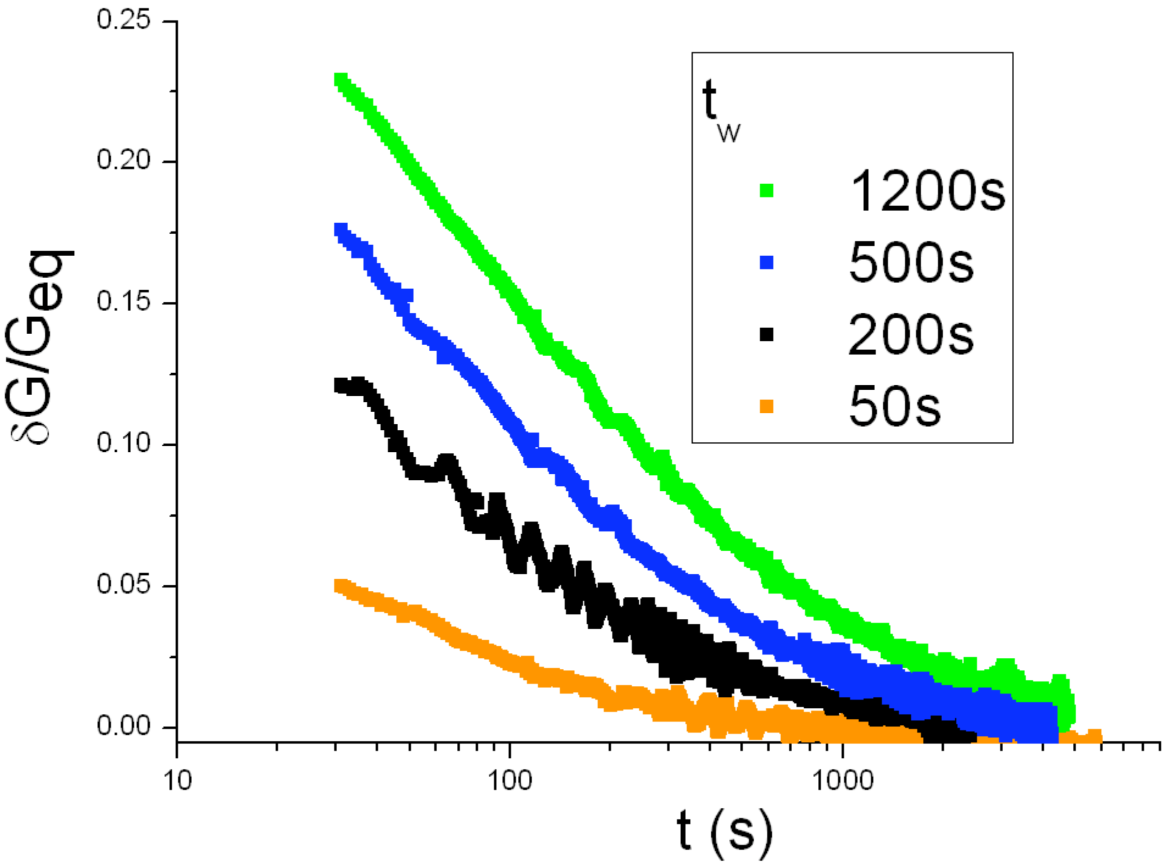


Also the "excitation" is in fact a relaxation toward a new quasi-equilibrium configuration

We study the variable *y*:

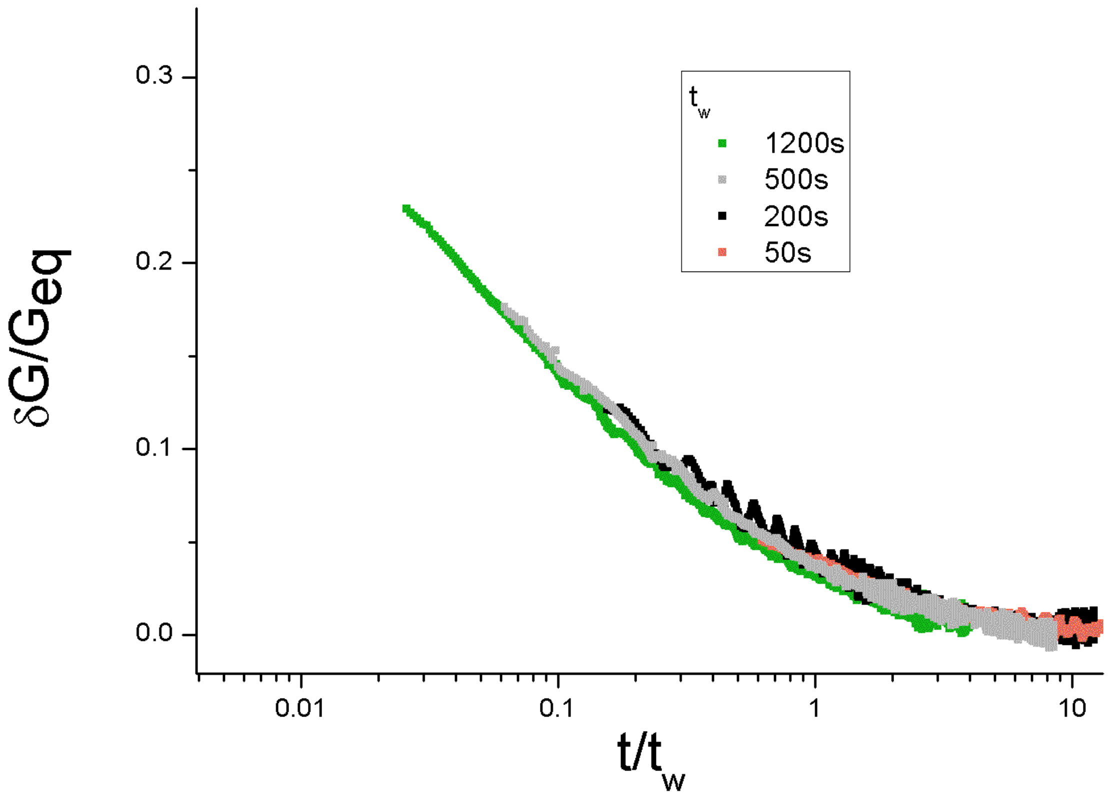
$$y = \frac{\delta G}{G_{eq}} = \frac{|G - G_{eq}|}{G_{eq}}$$

Logarithmic relaxation and aging



$$y = y(t, t_w) \sim \log(t, t_w)$$ for $t<t_w$

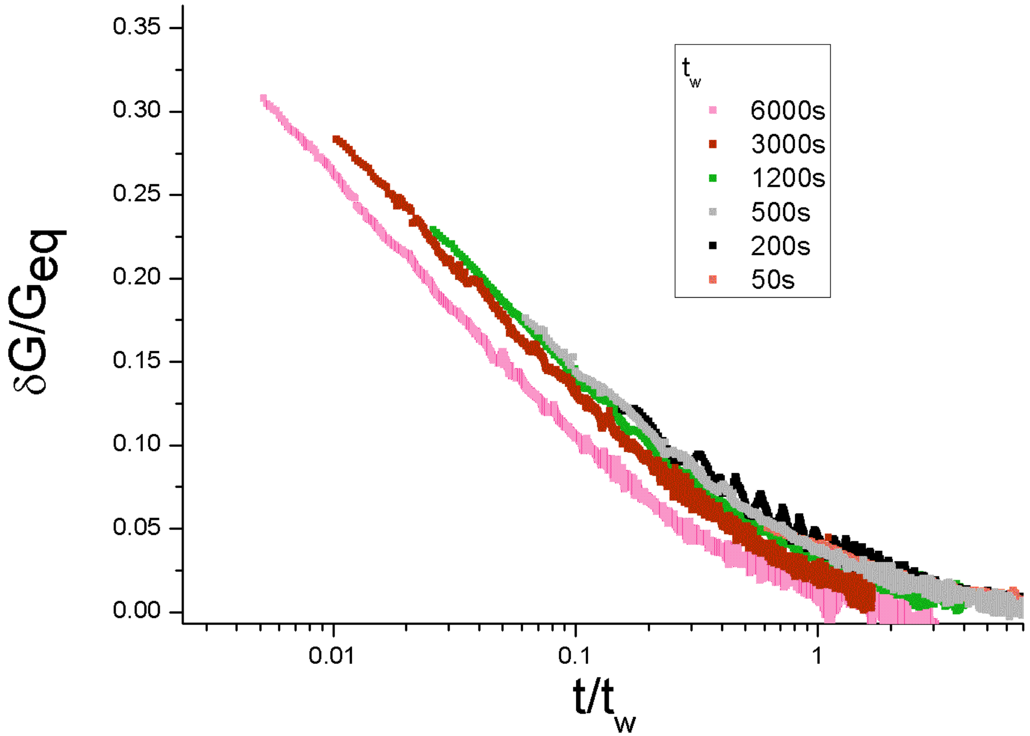
“Simple” aging: full scaling t/t_w



$$y(t, t_w) \sim \log(t / t_w)$$

“Simple” aging failure

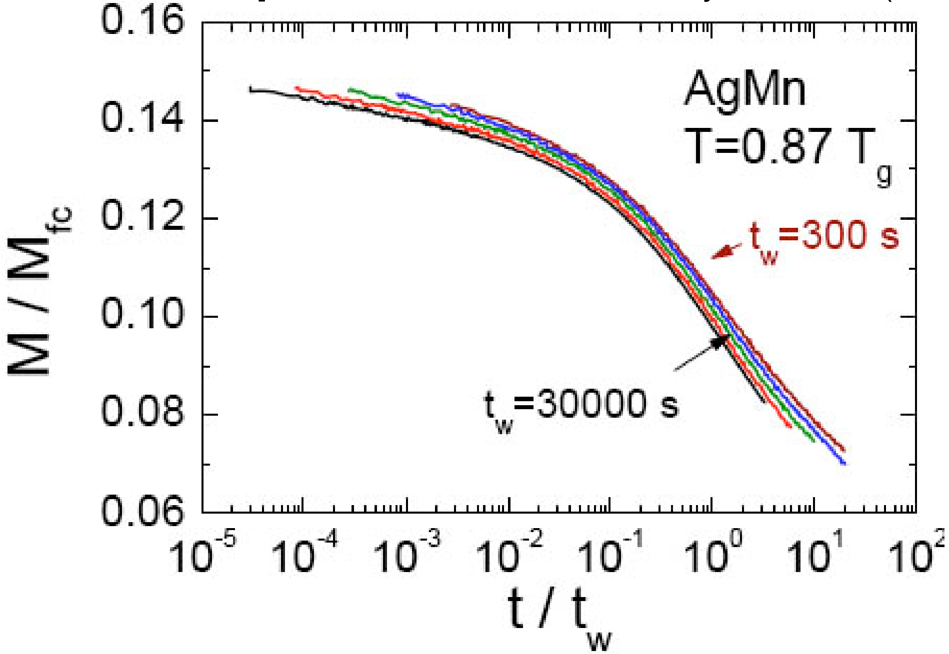
Beyond a given t_w value, a systematic gradual departure from the full scaling t/t_w is observed

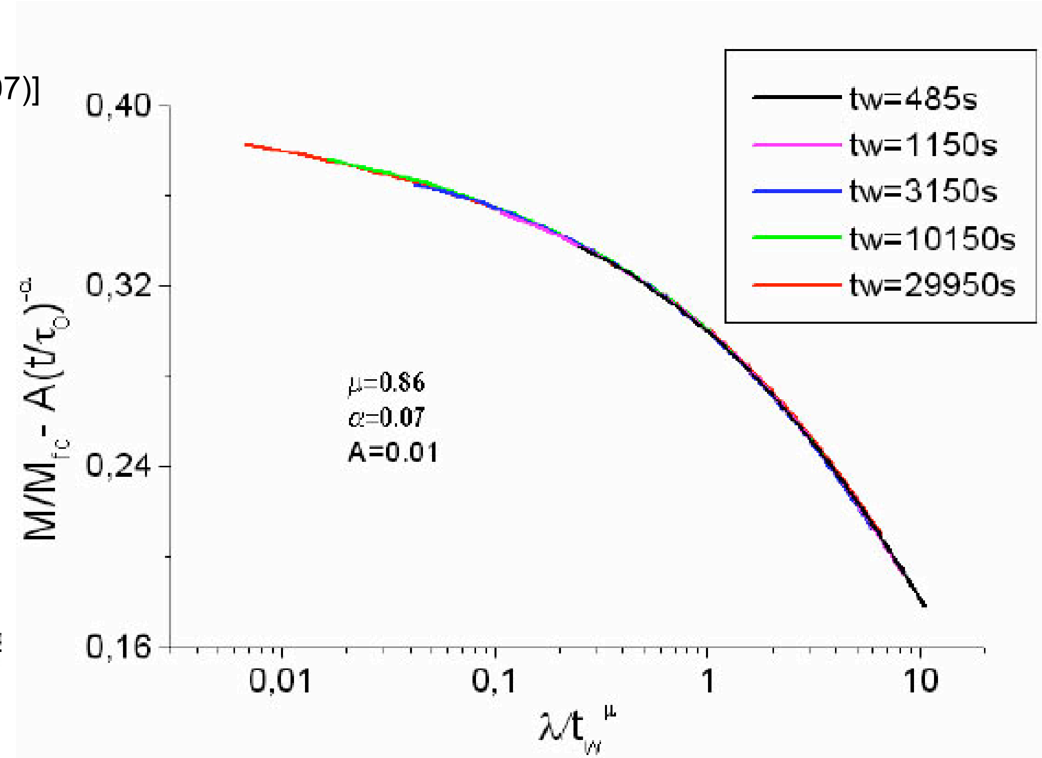


$$y(t,t_w) \neq f(t/t_w)$$

"Simple" aging failure: "subaging"

[E. Vincent, *Lecture notes in Physics* 716, 7 (2007)]



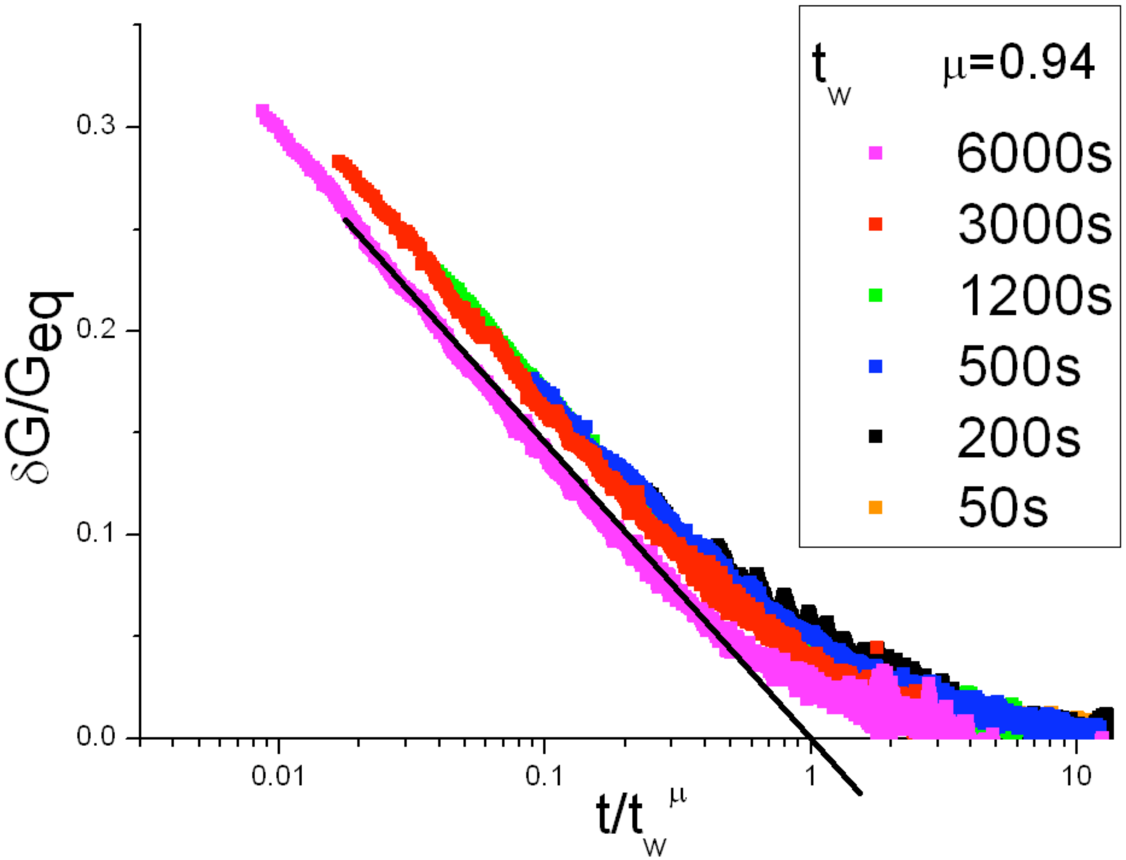


$$y(t, t_w) = y(t/t_w^{\mu}) \quad \mu \leq 1$$

Observed in structural glasses and spin glasses, but lack of universality in the values of μ

Unclear physical meaning of μ

“Simple” aging failure



$$y(t,t_w) \neq \log\left(t/t_w^{\mu}\right)$$

Master function for the slow relaxation

For a thermally activated process (relaxation rate $\lambda \approx e^{-\frac{E}{kT}}$) and a uniform distribution of energy barriers:

Broad distribution of relaxation rates: $P(\lambda)d\lambda = P(E)dE \Rightarrow \quad P(\lambda) \approx \frac{1}{\lambda}$

Weighted sum of decaying exponentials: $F(t) = \int_{\lambda_{min}}^{\lambda_{max}} P(\lambda) \cdot e^{-\lambda t} d\lambda = \int_{\lambda_{min}}^{\lambda_{max}} \frac{e^{-\lambda t}}{\lambda} d\lambda$

$1/\lambda_{min}$: characteristic time of the slow relaxation

$$F(\lambda_{min}, t) = \int_{\lambda_{min}}^{\infty} \frac{e^{-\lambda t}}{\lambda} d\lambda \equiv \mathrm{ExpInt}(\lambda_{min}, t)$$

$\approx -\gamma_E - \ln(\lambda_{min} t)$ when $t << 1/\lambda_{min}$

Ref.: Van der Ziel, Physica (1950)
Pollak and Ovadyahu, Phys. Stat. Sol. (c) 3, 283 (2006)

Master function and aging

Excitation: slow relaxation toward another metastable state: $\delta G^{exc}(t) \approx \int_{\lambda_{min}}^{\lambda_{max}} \frac{(1-e^{-\lambda t})}{\lambda} d\lambda$

After a time $t=t_w$ the system starts to relax back to the pristine metastable state:

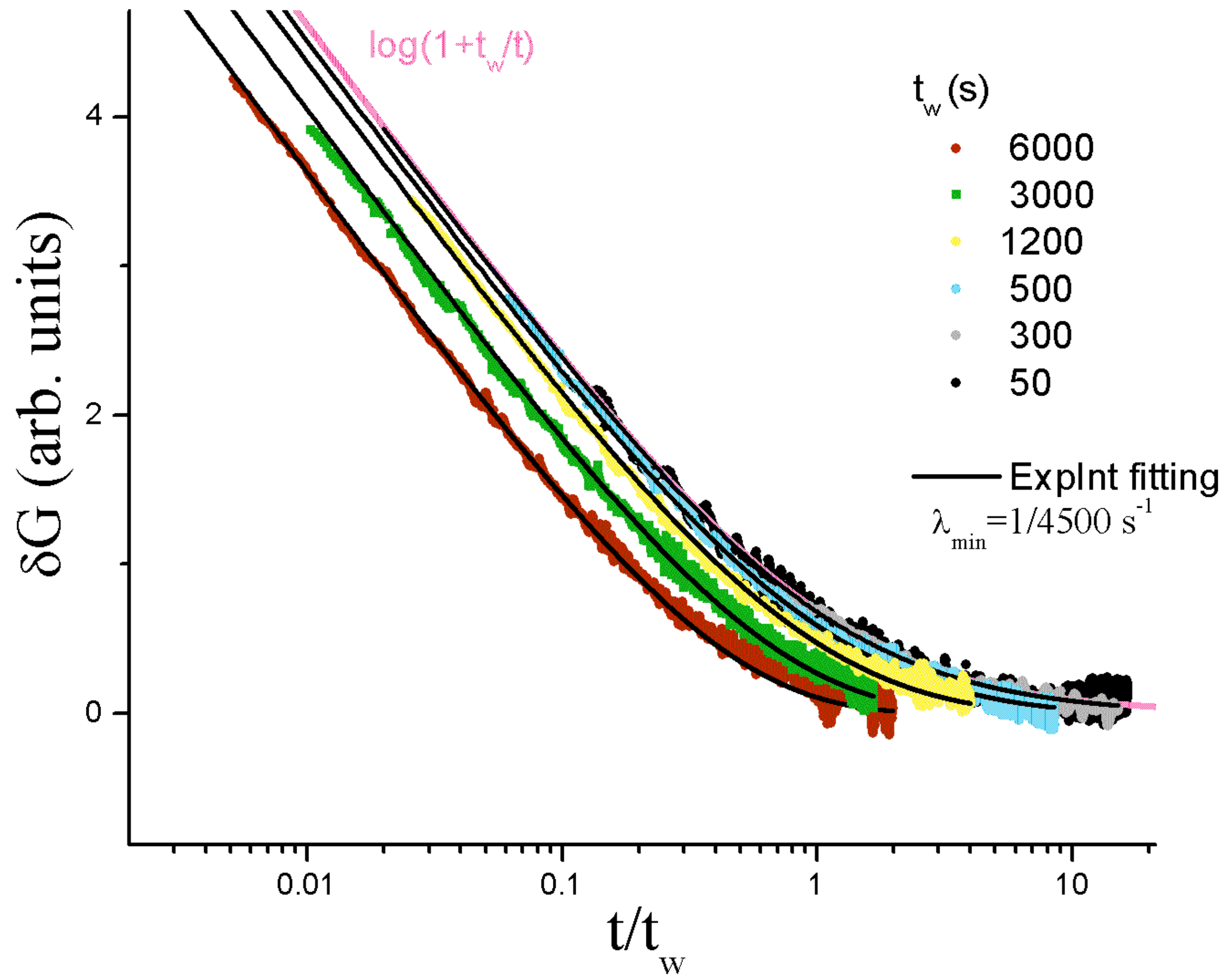
$$\delta G^{rel}(t,t_w) \approx \int_{\lambda_{min}}^{\lambda_{max}} \frac{(1-e^{-\lambda t_w})e^{-\lambda t}}{\lambda} d\lambda = F(t) - F(t+t_w)$$

If $F(\lambda_{min},t) \approx -\gamma_E - \ln(\lambda_{min} t) \quad \Rightarrow \delta G(t,t_w) = \delta G(t/t_w) \approx \log(1+\frac{t_w}{t})$

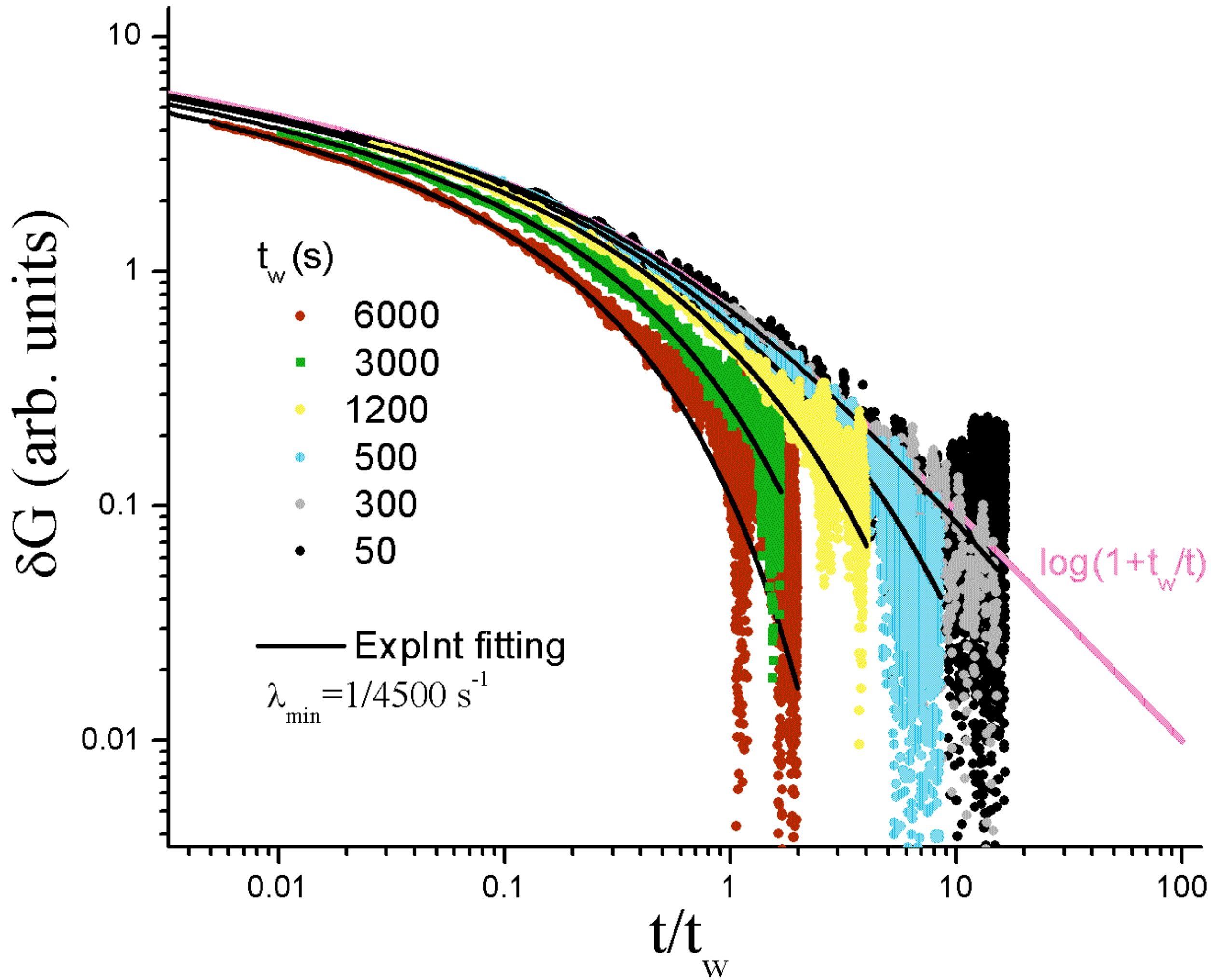
Full aging should be observed only if $t_w \ll 1/\lambda_{min}$ ⟵ Might be the reason for the observed full aging failure

Ref.: Amir et al, PRL 103, 126403 (2009)
Grenet, Eur. Phys. J. B 56, 183 (2007)

Fitting data: one parameter (λ_{min})

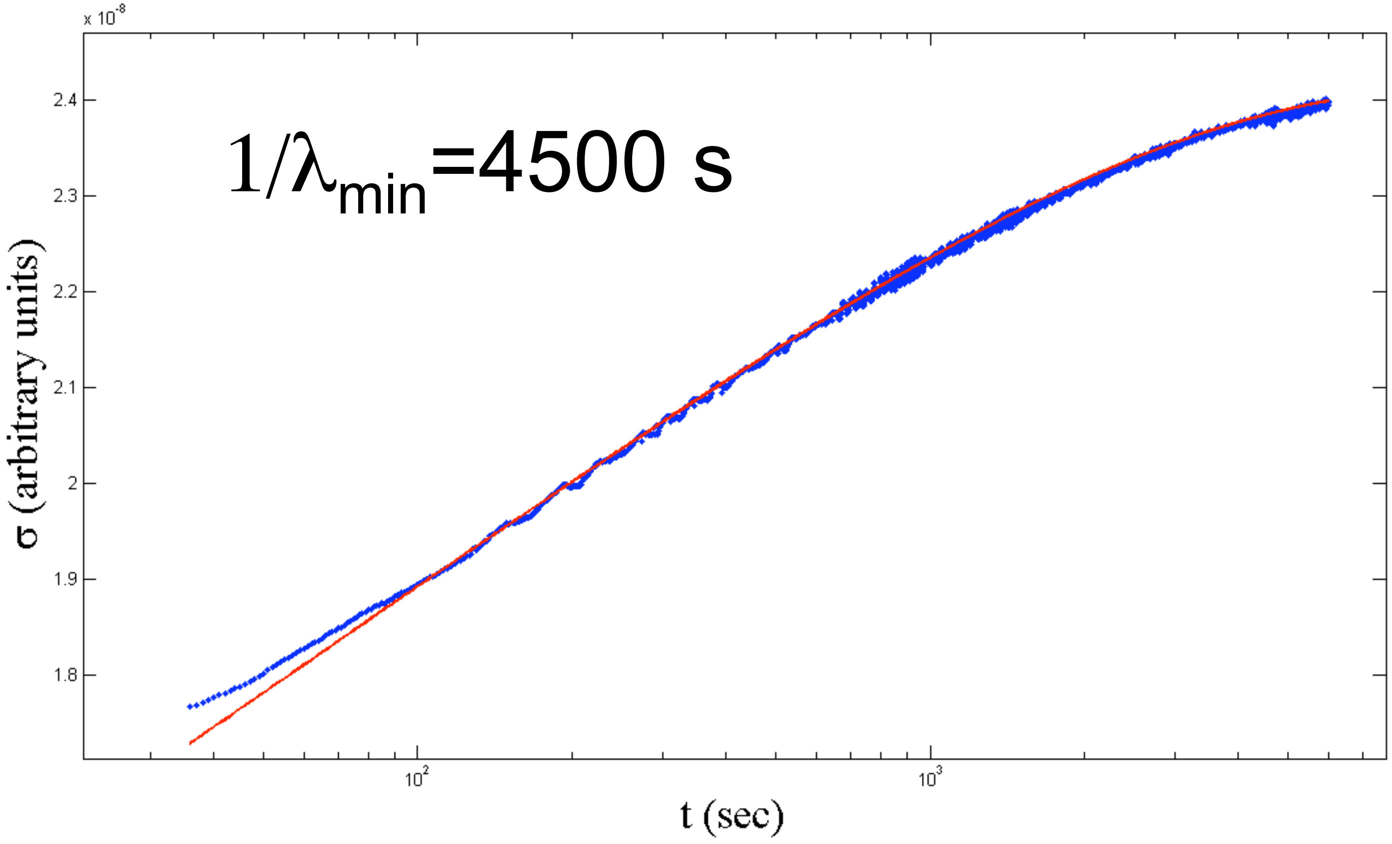


Fitting data: one parameter (λ_{min})

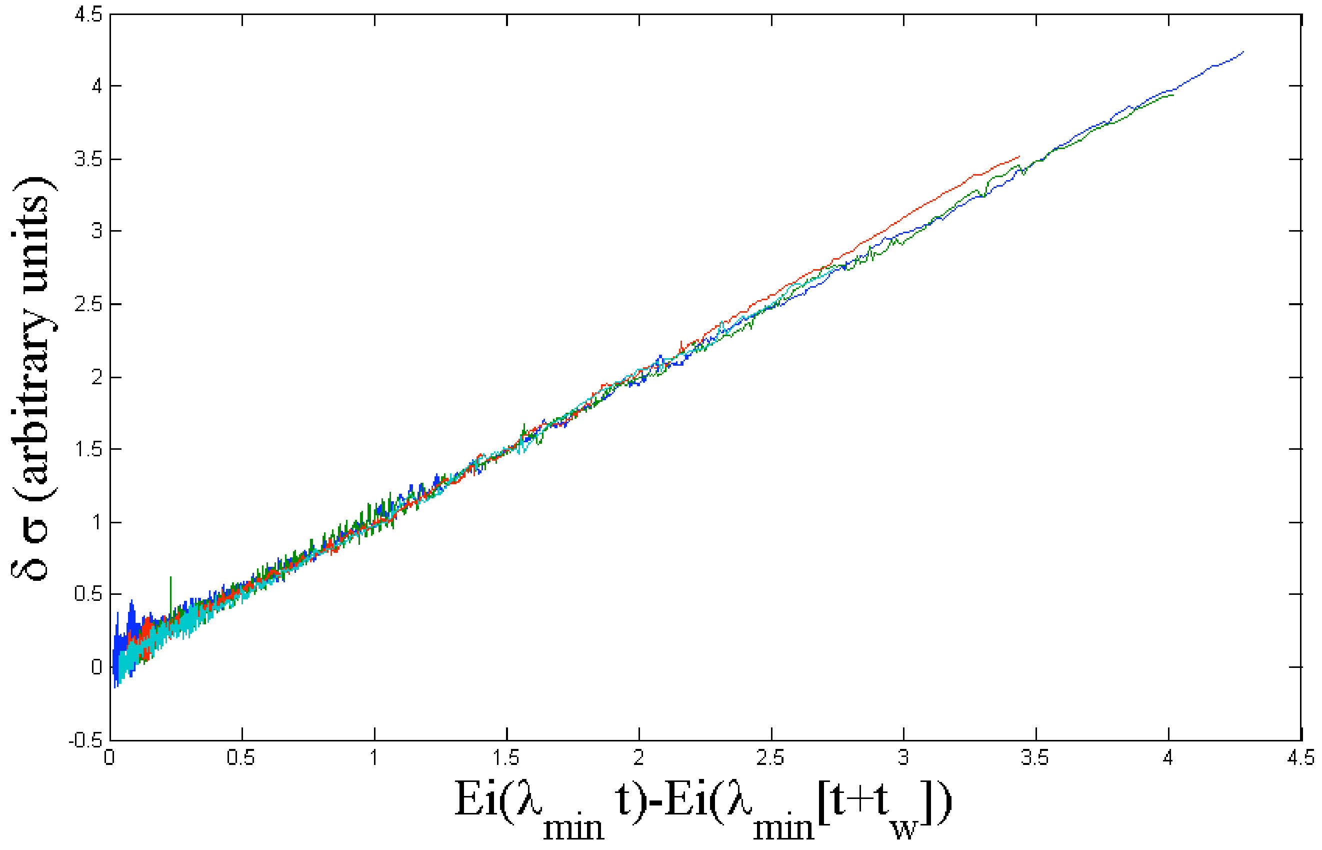


Fitting data: one parameter (λ_{min})

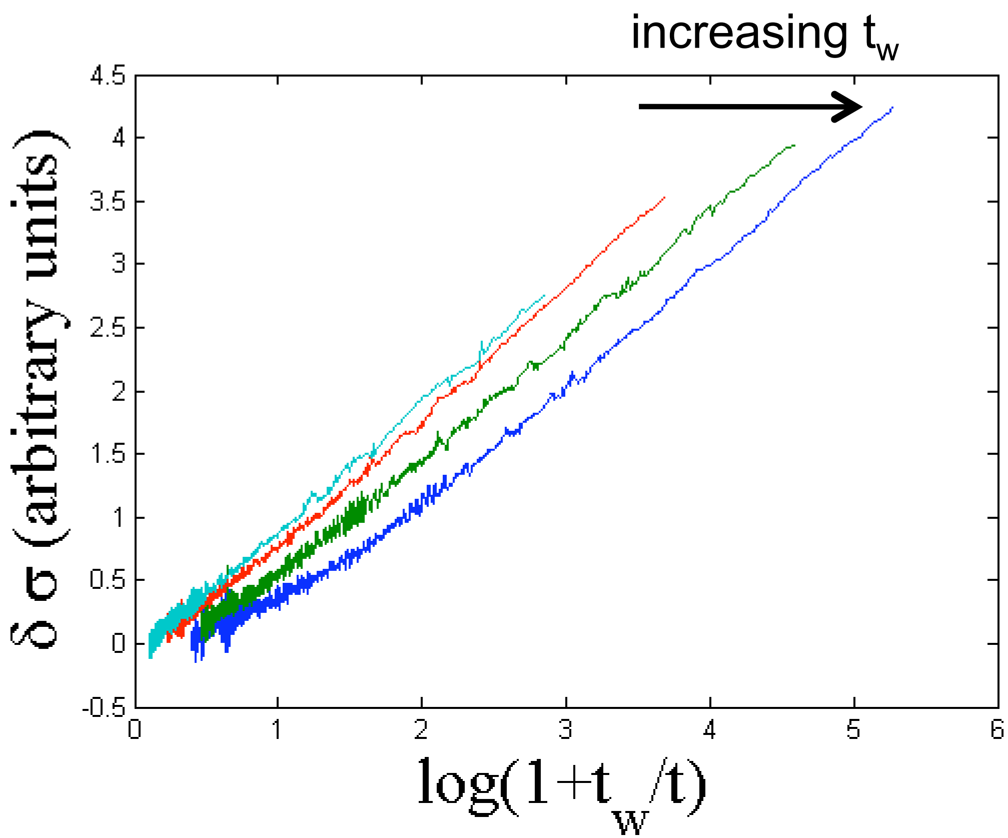
Excitation:



Fitting data: one parameter (λ_{min})



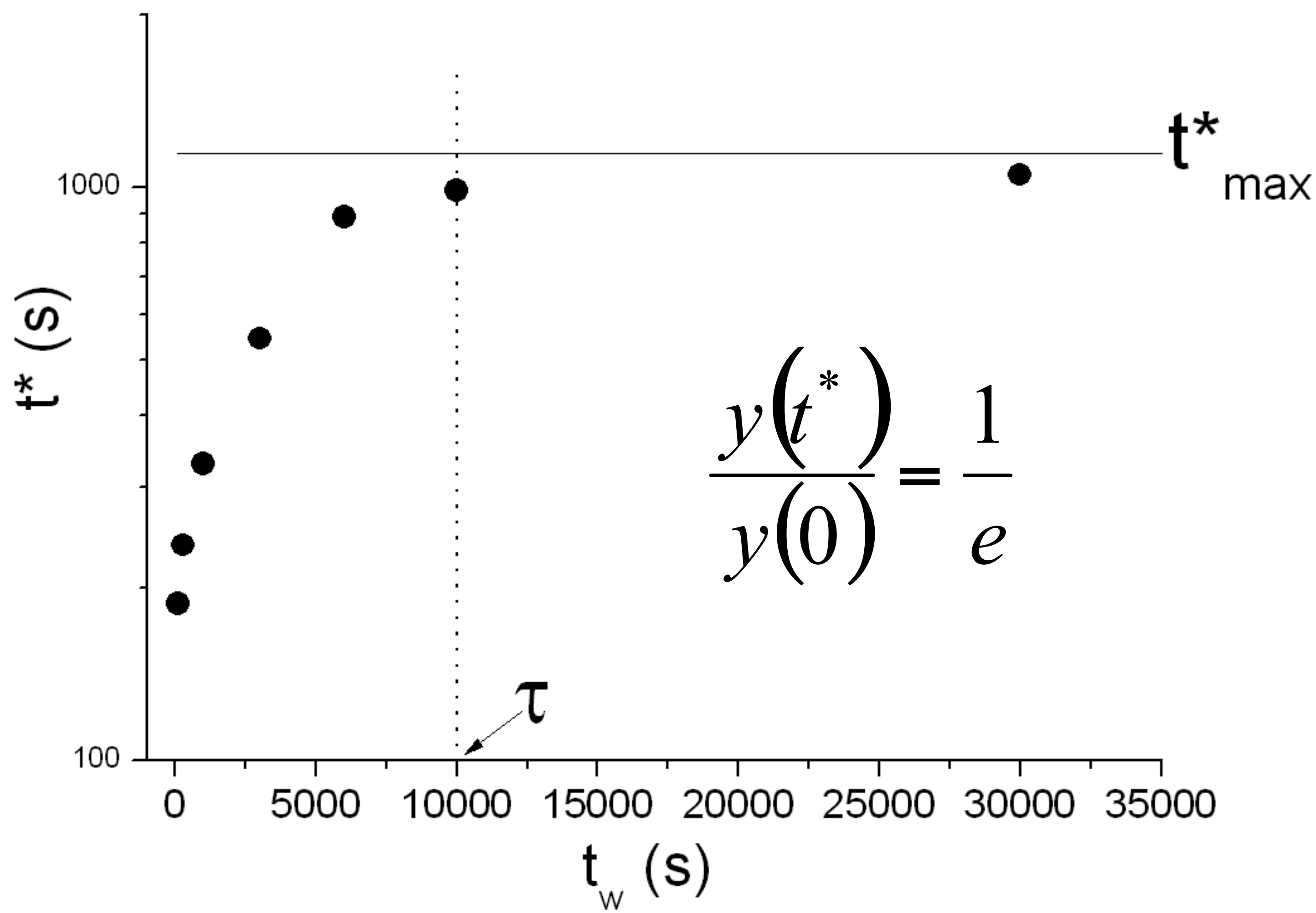
Failure of logarithmic fit and full aging



The departure from the logarithimic behavior (and from the full aging) is directly related to the value of $(\lambda_{min}t_w)$.

In this regime, we can experimentally estimate the intrinsic time of the system which rules the slow relaxation.

Relaxation time



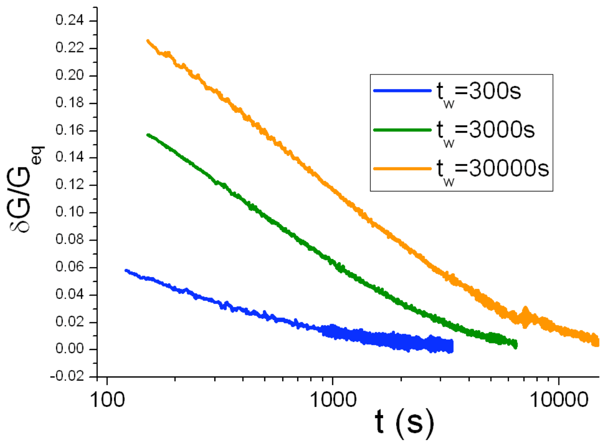
$$\frac{y(t^*)}{y(0)} = \frac{1}{e}$$

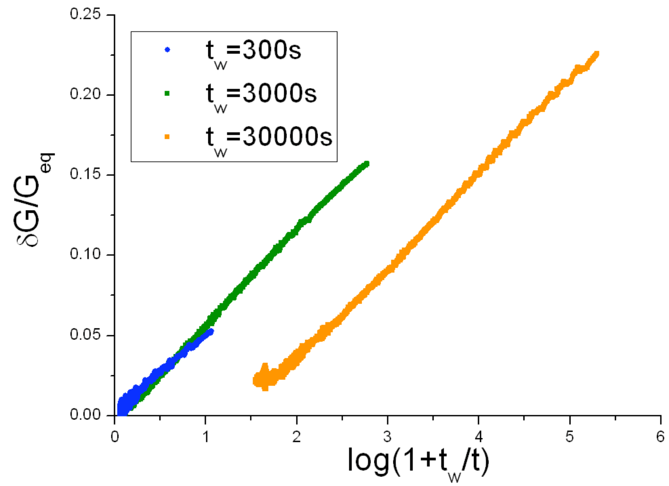
t* depends on arbitrary definitions.

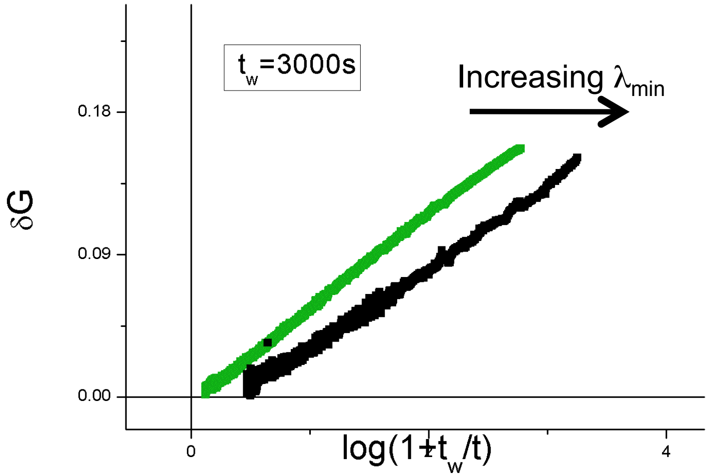
$\tau = 1/\lambda_{min}$ is an intrinsic property of the system

Failure of logarithmic fit and full aging

Same sample after 2 months in air



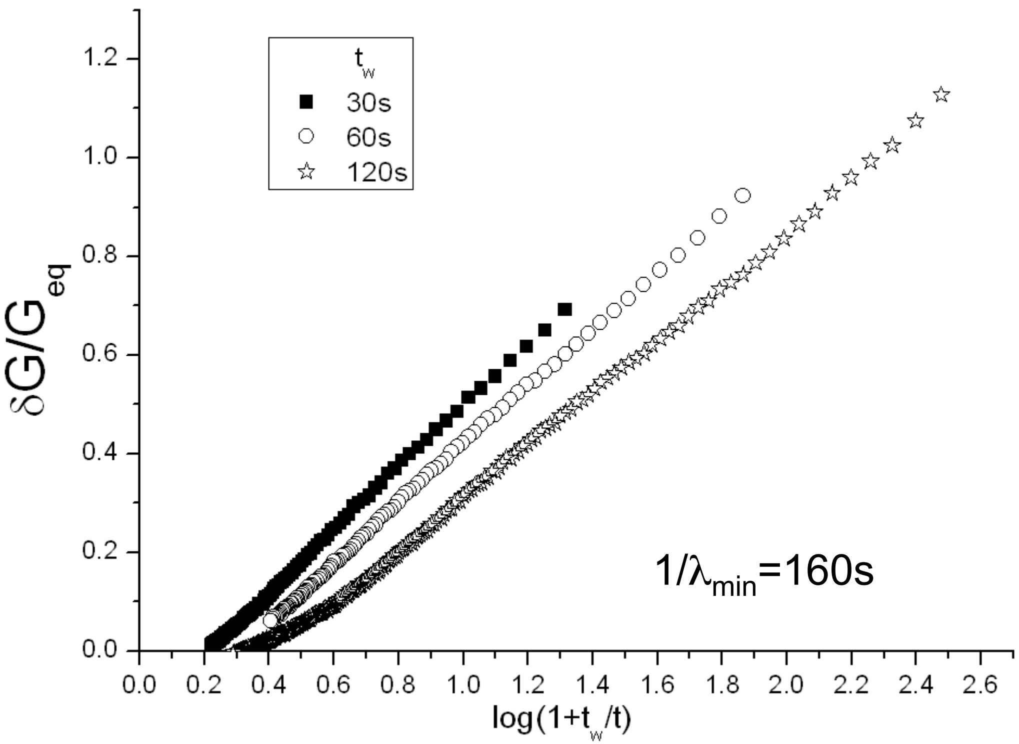




This system is slower than the previous one.
More traps are likely to be there

Dependence on traps density

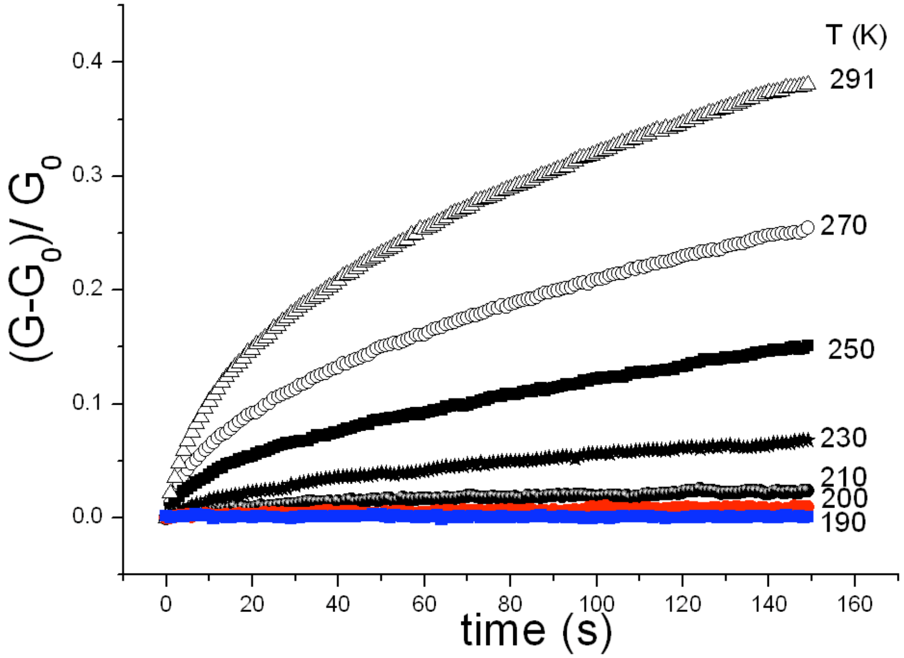
“fresh” sample (more free carriers, less traps)

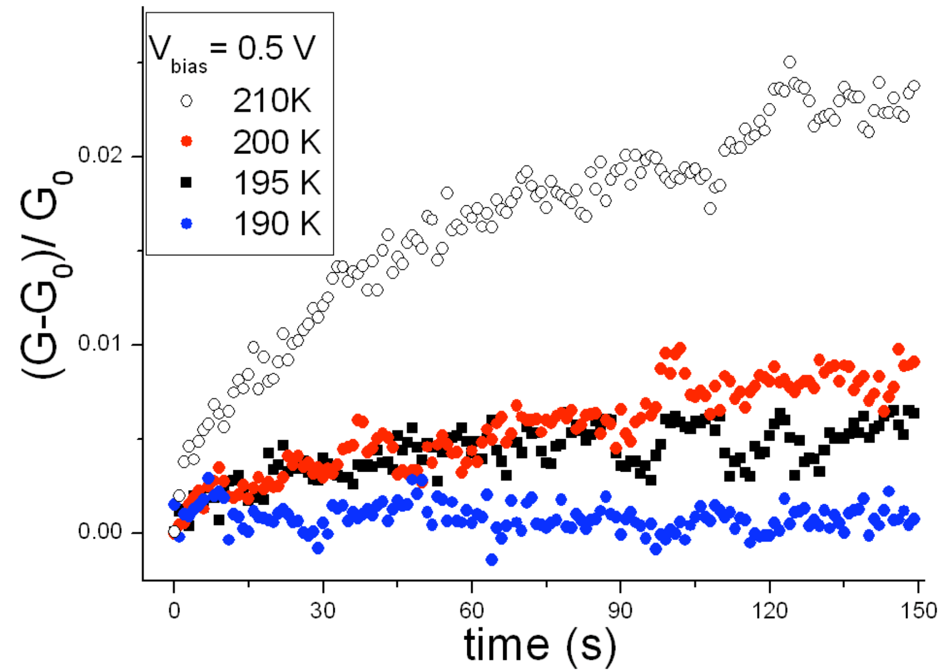


dynamics is faster in as-prepared samples

Effect of temperature

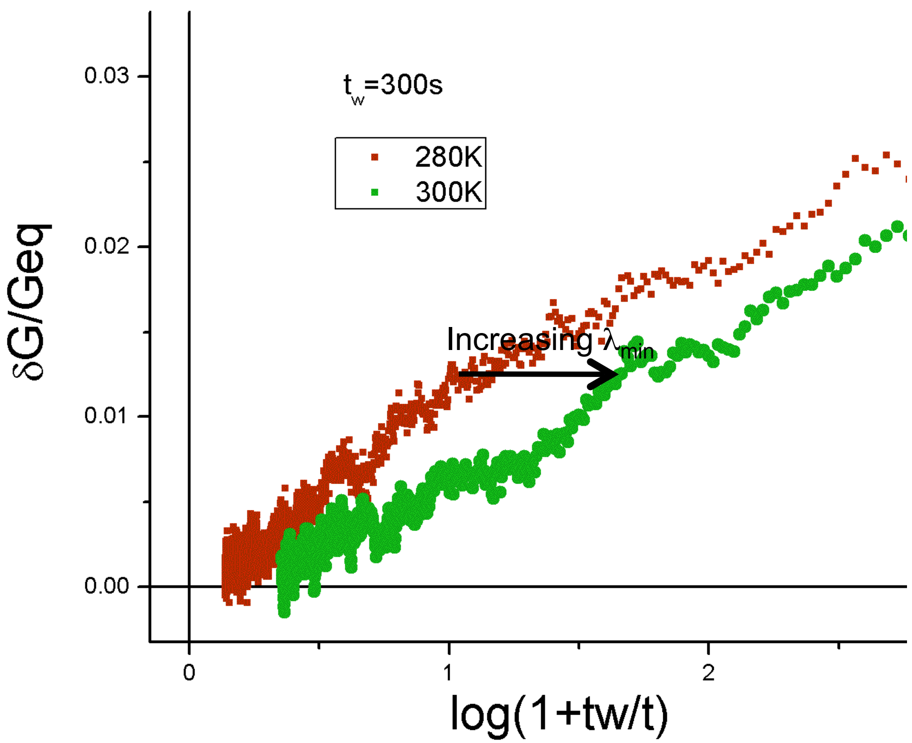
Excitation:

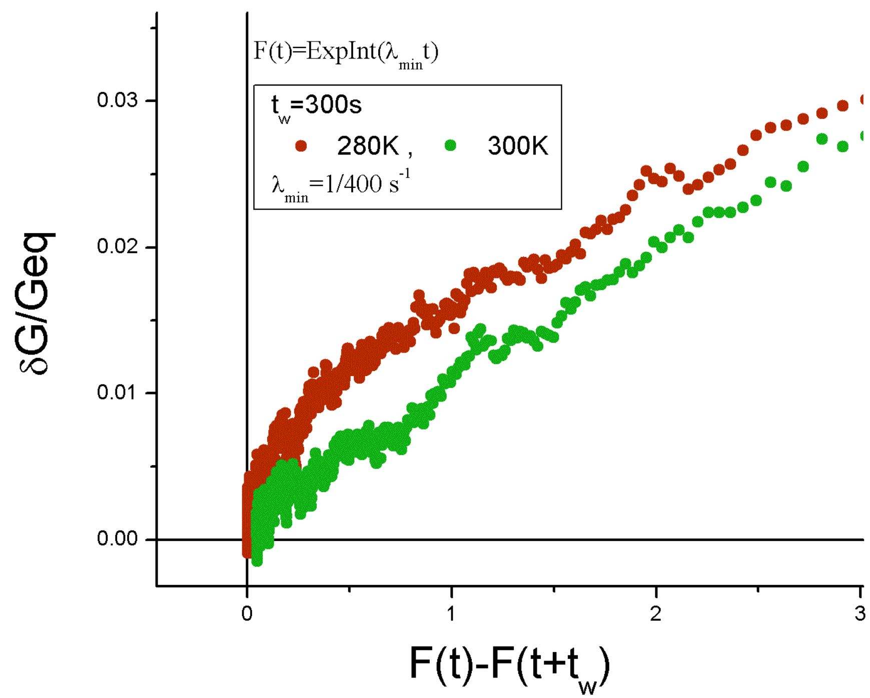




With cooling the sample, exciting the system out of equilibrium becomes harder and harder

Effect of temperature on the slow dynamics





Dynamics slow down upon cooling
Thermally activated dynamics?

Extrinsic electronic glass?

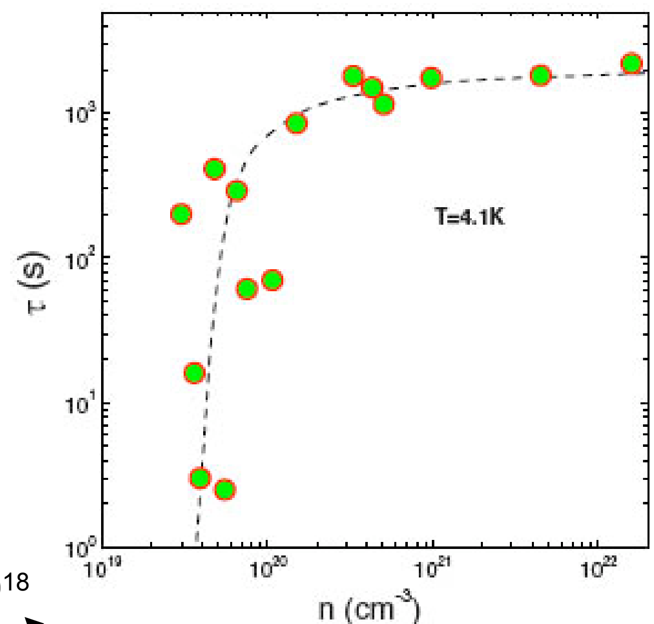
Qualitative differences with respect to InOx electron glass:

Dynamics slows down with decreasing T: quantum electron glass should be ruled out.

Moreover, we have systematically observed that more oxidized samples display smaller $1/\lambda_{min}$, i.e., the typical relaxation time τ increases with decreasing free carrier density, and with increasing the traps density

Can the dependence on density be an indication for correlated dynamics? (i.e. relaxation of non indipendent but interacting entities?)

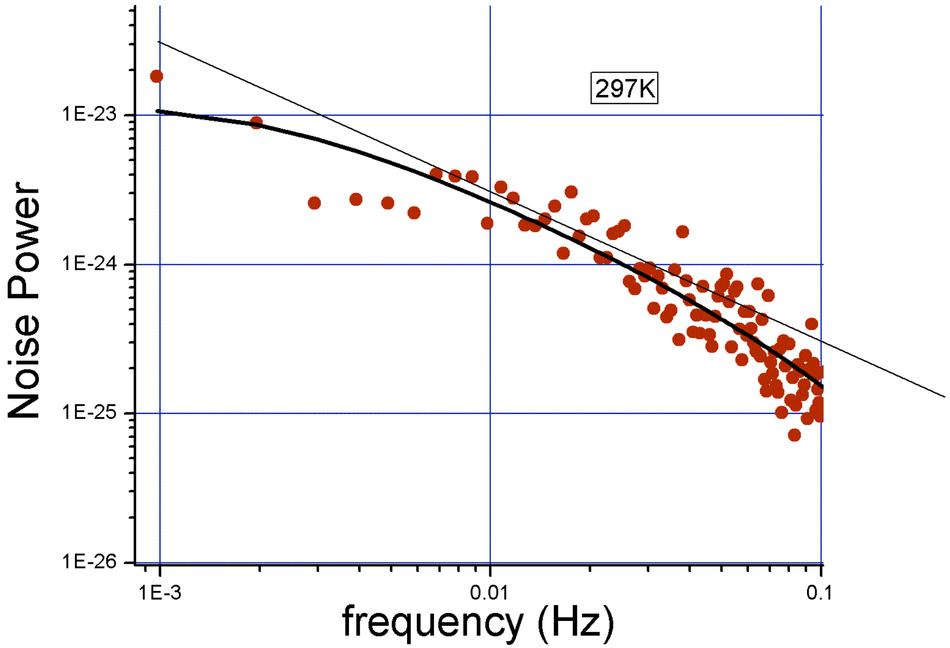
PHYSICAL REVIEW B 78, 195120 (2008)

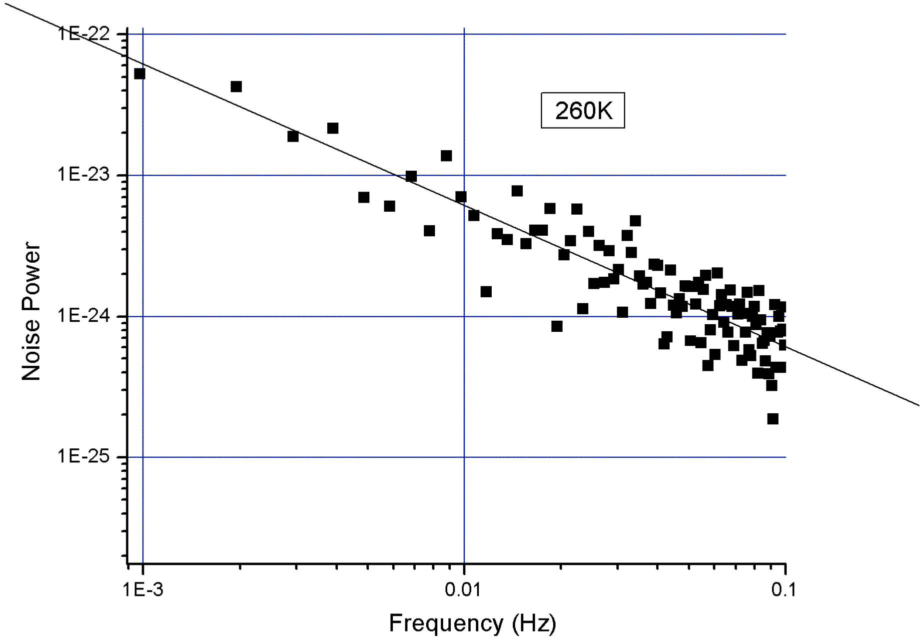


10^{18}

porous Si

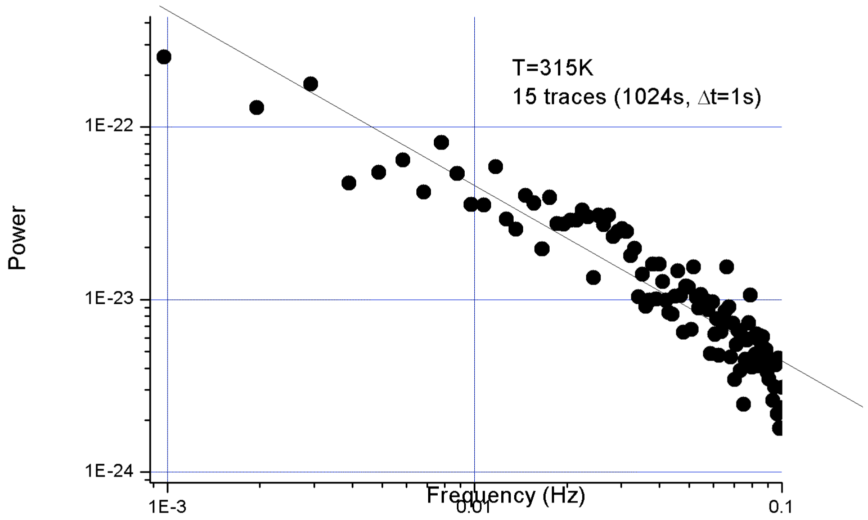
1/f noise - (preliminary results to be checked)

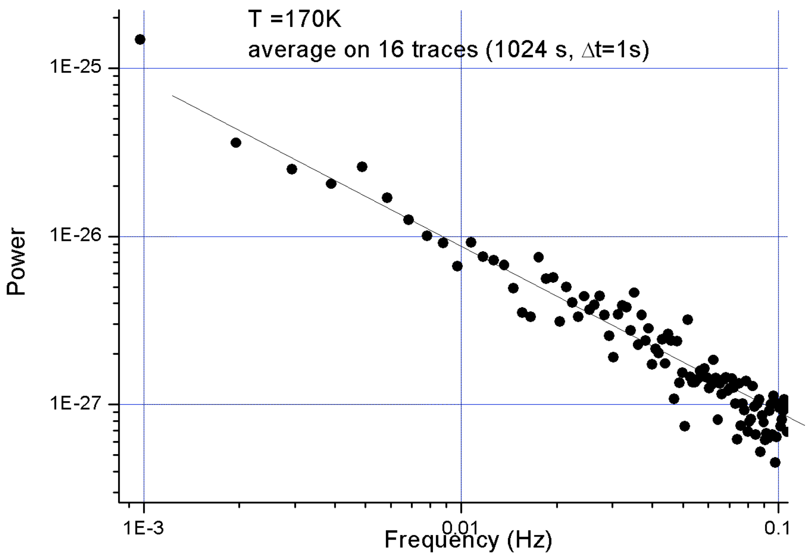


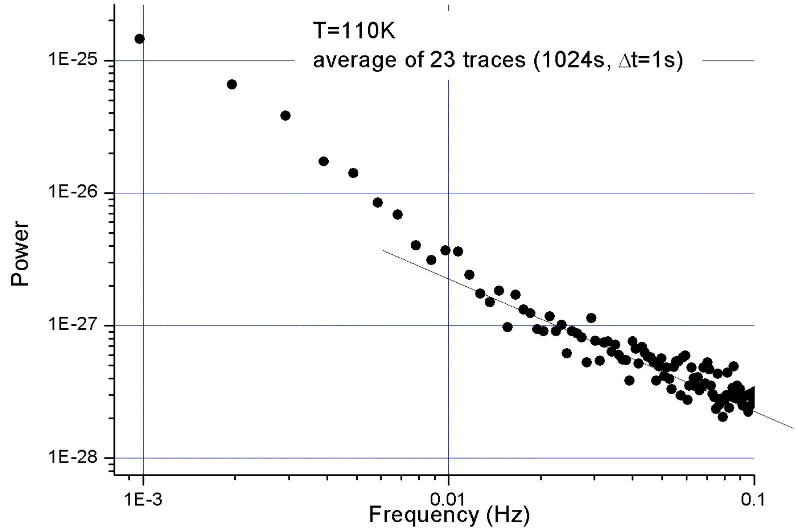


See: Amir et al., Ann. Phys. 2009

1/f noise - (preliminary results to be checked)



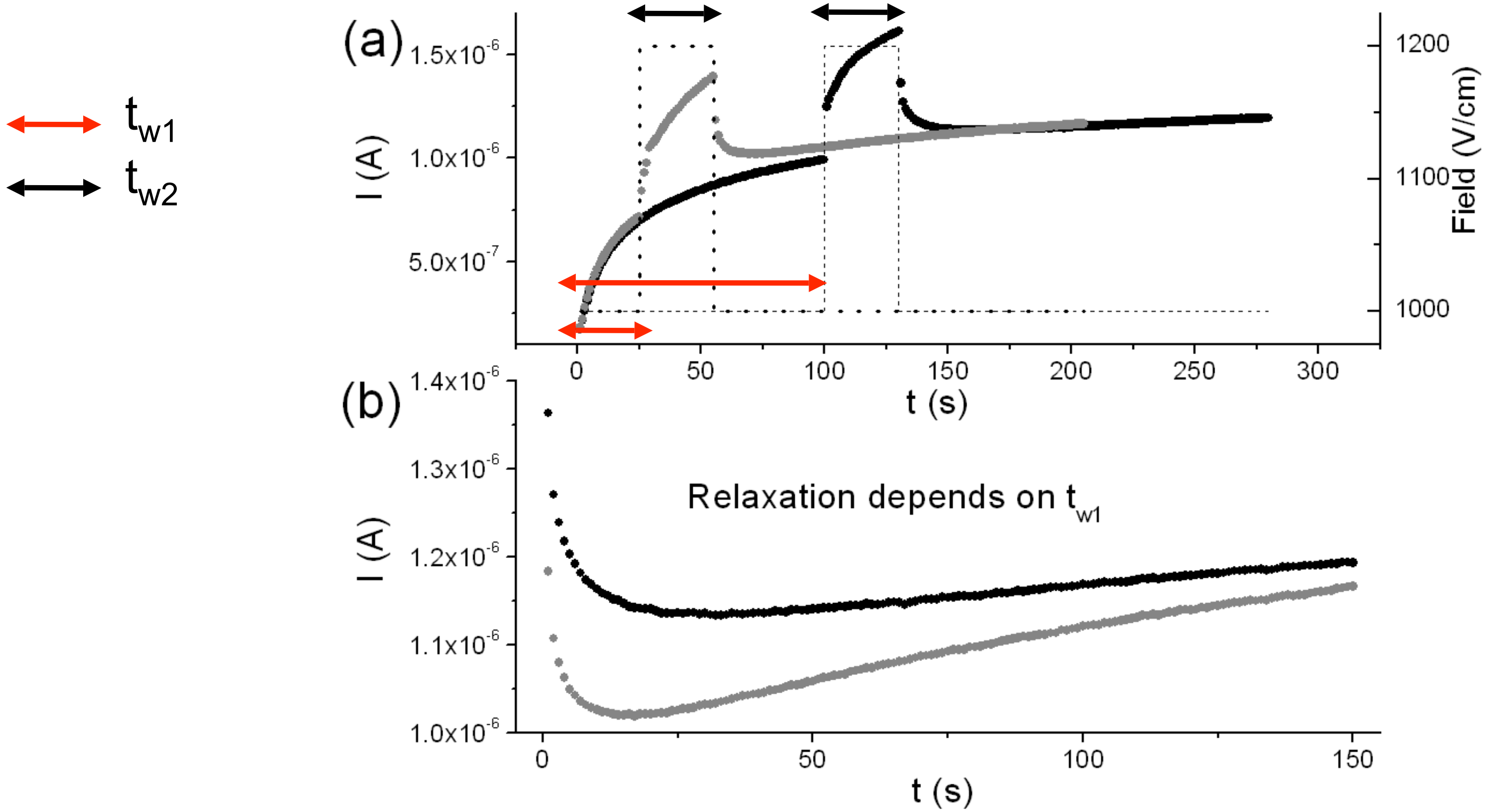




Other “glassy” effects

Age (hystory)-dependent dynamics: “memory”

Relaxation depends also on t_{w1}



Some conclusions

Slow relaxation and aging described by the exp integral ($\Leftarrow \mathrm{P}(\lambda) \approx \frac{1}{\lambda}$)

An intrinsic characteristic time of the system ($1/\lambda_{min}$) can be experimentally evaluated

Dynamics slows down with decreasing T and with increasing traps density

Possible study of glassy behavior at RT and by reproducible and precise electrical measurements:
Universal aspects of glasses may be tested