The first nanoseconds in the lifetime of a phase change material glass

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Germanium telluride as a fragile glass former

Germanium telluride (GeTe) is a phase change material and, therefor, a fragile glass former with a large contrast in electrical properties between the crystalline and amorphous phase.

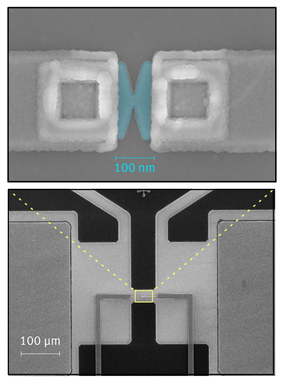
Phase change materials are of interest for applications such as (multibit) non-volatile data storage and as model systems for glass dynamics.

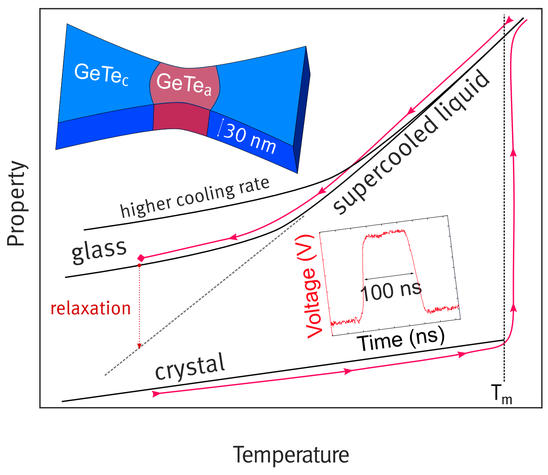
Switching the cell between crystalline and amorphous phase:

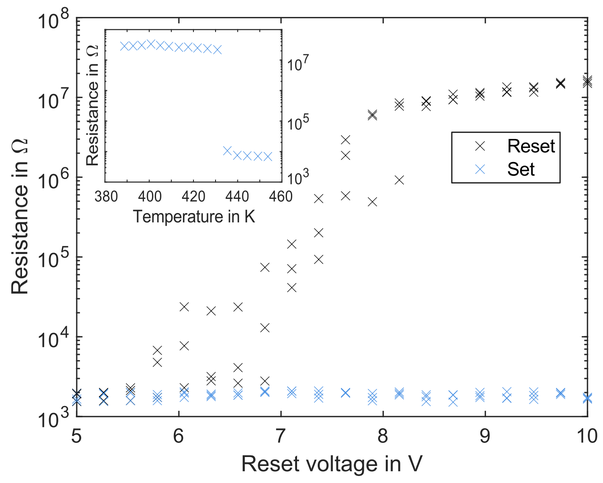
- to avoid crystallisation: high quenching rates (~ 10^9 K/s) necessary → nanoscopic volumes
- melting the material using 100 ns voltage pulses of varying amplitude leads to different amorphous volumes
- recrystallisation using the platinum micro heater underneath the GeTe cell
- the cell can be switched back and forth at least ten thousand times

Relaxation of the GeTe glass:

- upon cooling the supercooled liquid the system falls out of equilibrium
- the formed glass relaxes towards this equilibrium, resulting in time dependent properties







Femtosecond pulse optical amorphisation

Instead of using nanosecond voltage pulses, femtosecond laser pulses can be used to amorphise the material. This enables measurements of the resistance down to nanoseconds and reveals deviations from the power law dependence of the resistance on time.

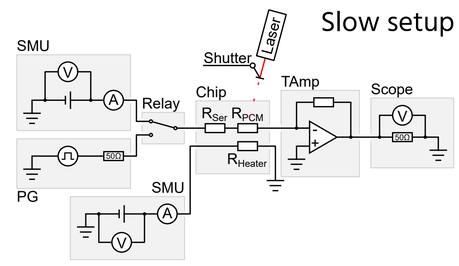
Resistance drift over 12 orders of magnitude in time

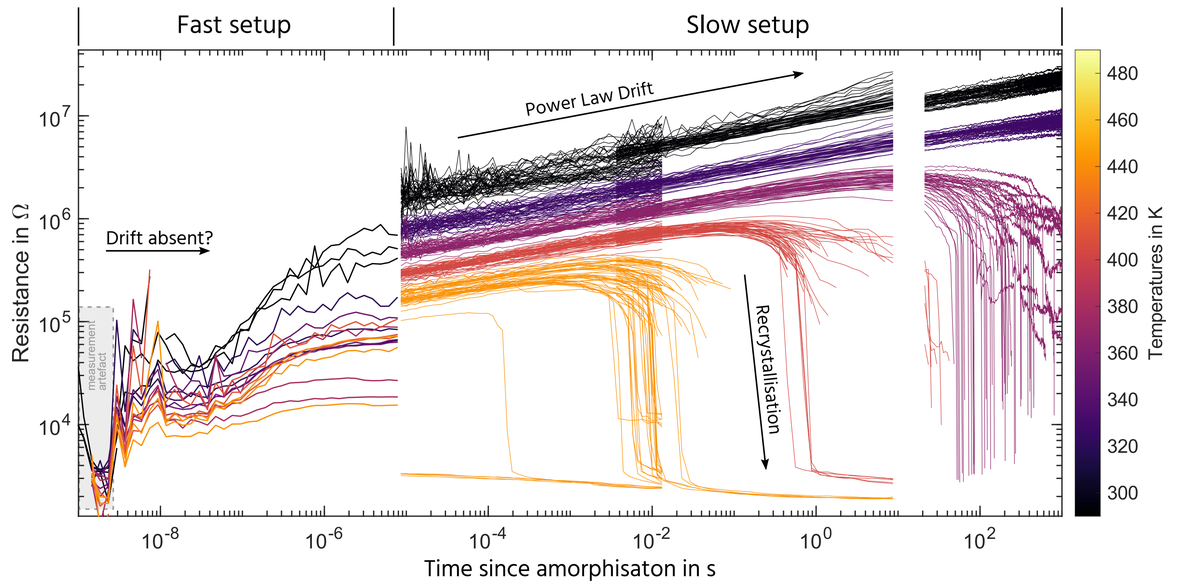
Amorphisation of GeTe using **laser pulses**

- pulse length of 60 fs
- wavelength of 1550 nm for selective absorption in GeTe

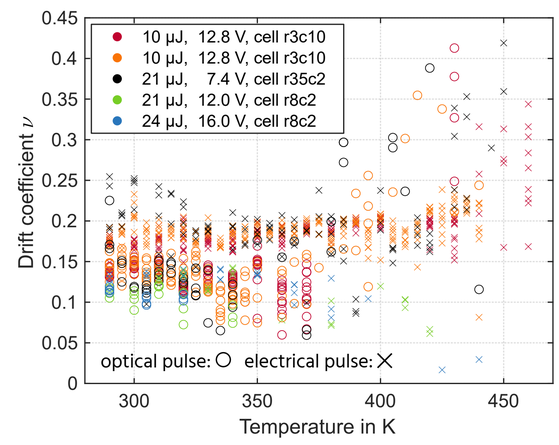
Two **deviations from power law** on short timescales

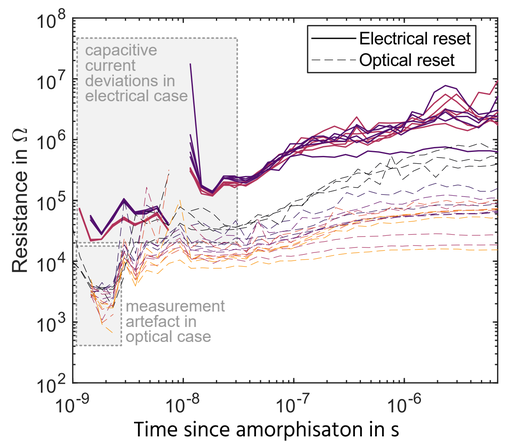
- drift absent for first 40 ns: Minimum energy barrier to overcome as first relaxation step?
- fast resistance increase afterwards: recombination of free charge carriers (as described in [2])?





Elecrical versus optical amorphisation





Resistance drift upon electrical amorphisation

Resistance drift describes the resistance increase of phase change material glasses over time. This presents a challenge for multi-bit storage applications.

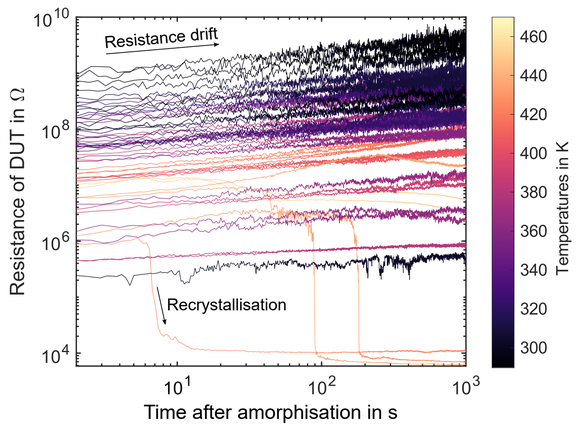
The mechanism behind drift is not well understood. Looking beyond usually considered measurement regimes might help to decipher the mechanism.

Resistance of phase change material glasses increases with a power law:

$$R = R_0 \cdot \left(\frac{t}{t_0}\right)^{\nu}$$
$$\Leftrightarrow$$
$$\log(R) = \log(R_0) + \nu \cdot \log\left(\frac{t}{t_0}\right)$$

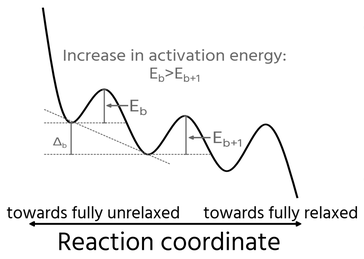
- in a log-log plot: straight line with drift coefficient ν as the slope
- at larger temperatures: recrystallisation

The resistance drift could be linked to **structural relaxation** as suggested in the following model:



Collective structural relaxation model [1]

- Unrelaxed glass differs from the relaxed supercooled liquid by a finite number of structural defects
- Every relaxation step leads to a local stabilisation: incresing activation energy for subsequent relaxation steps



- linear increase in activation energy to remove defects leads to logarithmic relaxation with time
- with multiple trapping model: power law increase of resistance over time

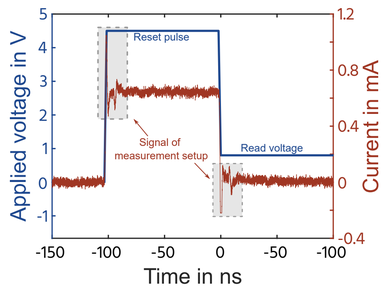
Measurements on nanosecond timescales - Capacitative currents

In the moment of amorphisation the applied voltage has to be reduced as fast as possible.

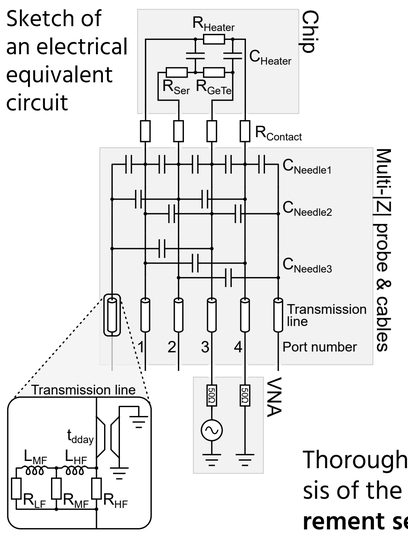
$$I(t) = C \cdot \frac{\partial U(t)}{\partial t}$$

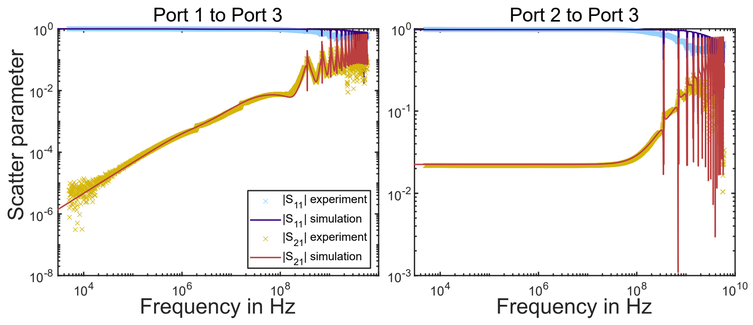
Current on short timescales dominated by capacitive currents.

This makes resistance measurement on these short timescales very challenging!



Sketch of an electrical equivalent circuit



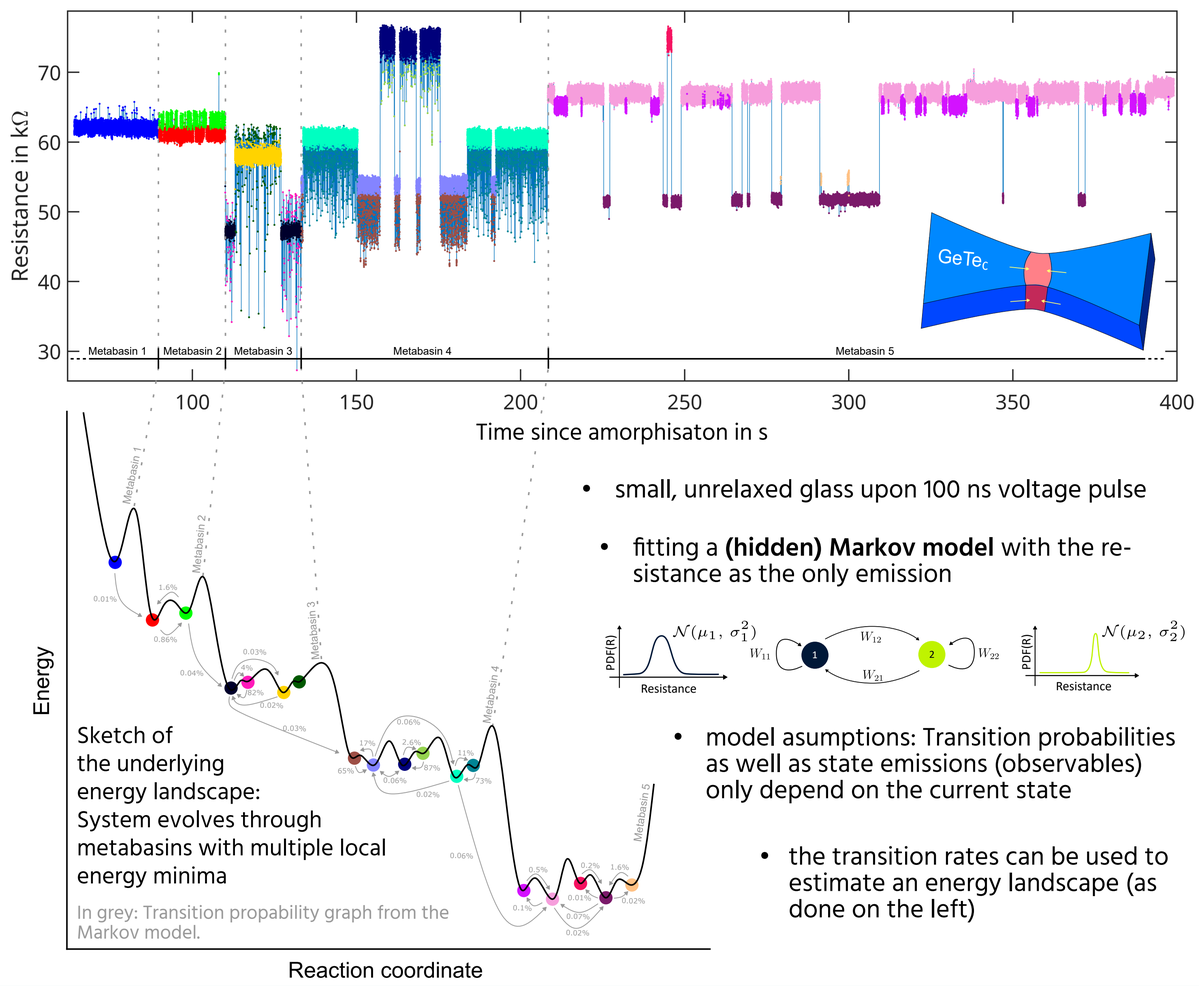


Thorough analysis of the **measurement setup**:

- measurements of the scatter parameters through different ports
- fit the model on the left to get electrical equivalent circuit

Discrete relaxation steps in small volumes

Reducing the volume of amorphous phase change material reveals a different deviaton from the power law: fluctuations of the resistance between dis-crete levels. This could be explained by the glass moving through the energy landscape. Being highly sensitive to the microstucture, the electric resisistance can be used as an observable to analyse this energy landscape.



Sketch of the underlying energy landscape: System evolves through metabasins with multiple local energy minima

- small, unrelaxed glass upon 100 ns voltage pulse
- fitting a **(hidden) Markov model** with the resistance as the only emission
- model asumptions: Transition probabilities as well as state emissions (observables) only depend on the current state
- the transition rates can be used to estimate an energy landscape (as done on the left)

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[1] Manuel Le Gallo et al, Collective structural relaxation in phase-change memory devices. Advanced Electronic Materials, 4(9), 2018
[2] Daniele Ielmini et al, Recovery and drift dynamics of resistance and threshold voltages in phase-change memories. IEEE Transactions on Electron Devices, 54(2), 2007