Analyse et simulations de modèles issus de la nano électronique

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Introduction

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1. Introduction
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The increasing use of portable electronic devices that require data storage implies a strong need for non-volatile memories.

This type of memory is to store information when power is off.

The objective of the industrial thus to decrease in size the memory cells to enable the user to store more items.
Most known: Flash memory

- Flash memory of a form of semiconductor memory is widely used for many electronics data storage applications.
- Dates back to around 1980 developed at Toshiba by Fujio Masuoka.
- Today in addition to this flash memory storage (flash memory USB memory sticks, digital camera memory cards in the form of compact flash or secure digital, SD memory) is used in many other items from MP3 players to mobile phones, and in many other applications.
Problem: Downscaling Flash memory seems to have reached its limits
Solution: Research is conducted on new types of memory

Resistive type memory
- The information is stored as a resistive state of a metal-insulator-metal structure
- These memories have a state of high resistivity and one of low resistivity and you can control the transition from one state to another ("Resistive Switching").
- The state on corresponds to low resistivity and the state off to the high resistivity. SET is called passing the off state to the on state and the reverse phenomenon is called RESET.

Properties required
- The difference in resistance between the off state and the on state must be meaningful in order to differentiate the two states
- The cell must withstand at least $10^7$ on-off cycles
- A retention period of at least 10 years as flash memory.
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Introduction

**Figure:** Memory Taxonomy
CBRAM (Conductive-Bridge Random-Access memory)

- Promising technology due to
  - Low operating voltages
  - Low power consumption
  - Ease of integration in the back end of a logic process
- Is composed of a resistive switching layer (GeS2, GeS)
- An electrochemically active electrode (Silver, Copper)
- An electrochemically inert counter electrode (Platinum or Tungsten).

**Figure:** CBRAM cell - LETI CEA Grenoble
**Figure:** Optical microscopy image of a Ag dendrite grown from the (−)Au electrode towards to (+)Ag electrode within a $AS_2S_3$ thin film on a glass substrate.
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The model: physical aspects

Under a positive voltage on the active electrode (SET)

(i) Anodic dissolution by Ag oxidation:

\[ Ag \rightarrow Ag^+ + e^- \]

(ii) Migration of the \( Ag^+ \) cations through the solid electrolyte under the action of the electric field and the gradient of concentration.

(iv) Filament growth through the reduction of \( Ag^+ \) ions on filament surface according to the reaction:

\[ Ag^+ + e^- \rightarrow Ag. \]
Mathematical model

The model

Oxidation and reduction reaction are modeled with Butler-Volmer equation

\[ J_{BV} = j_0 \left( \exp \left( \frac{\alpha e \eta_{B_1}}{k_B T} \right) - \exp \left( \frac{-(1 - \alpha) e \eta_{B_1}}{k_B T} \right) \right) \]

- \( j_0 \) is a exchange current density
- \( k_B \) is the Boltzmann constant, \( T \) is the temperature, \( e \) is the elementary charge.
- The over-voltage \( \eta_{B_1} \)
- A charge transfer coefficient \( \alpha \)

**Figure:** Schematic 2D view of a CBRAM cell
**Nucleation model** for the initialization of the filament (Milchev.)

- The nucleation rate is modeled through the equation:
  \[
  J_{\text{nuc}} = C(Z_0, N_{\text{crit}}) \exp\left(\frac{N_{\text{crit}} e |\eta_{\text{nuc}}|}{2k_B T}\right)
  \]
  \(\eta_{\text{nuc}}\) is the electrochemical overpotential.
  
- \(C(Z_0, N_{\text{crit}})\) depends on the nucleation sites number density \(Z_0\) and the number of atoms in a critical nucleus \(N_{\text{crit}}\)

**Figure:** Schematic 2D view of a CBRAM cell
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Mathematical model

The model

Hypothesis: space charge is reduced to $Ag^+$ cations

\[
\begin{align*}
\frac{\partial C_{Ag^+}}{\partial t} - \text{div}(eD\nabla C_{Ag^+} + e\mu C_{Ag^+}\nabla V) &= f_{1bd} \\
-(eD\nabla C_{Ag^+} + e\mu C_{Ag^+}\nabla V).\vec{n} &= J_{BV} \quad \text{on } B_1 \\
-(eD\nabla C_{Ag^+} + e\mu C_{Ag^+}\nabla V).\vec{n} &= J'_{BV} \quad \text{on } B_2 \\
-(eD\nabla C_{Ag^+} + e\mu C_{Ag^+}\nabla V).\vec{n} &= 0 \quad \text{on } B_3 \\
\end{align*}
\]

\[-\text{div}(\epsilon \nabla V) = eC_{Ag^+}\]

\[
\begin{align*}
V &= V_{app} \quad \text{on } B_1 \\
V &= 0 \quad \text{on } B_2 \\
\nabla V.\vec{n} &= 0 \quad \text{on } B_3
\end{align*}
\]

where $\epsilon$, $\mu$ and $D$ are the permittivity, $Ag^+$ ions mobility and the coefficient of diffusion of $Ag^+$
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We use a level set method (LSM) to simulate the growth of the filament.

- LSM introduced by Osher and Sethian '88 is used in many problems with an interface motion.
- The method implies a space-time function $\varphi$ with output between 0 and 1.
- The interface between filament and electrolyte is defined by the level set $\{\varphi = 0.5\}$.
- The level set function $\varphi$ is solution of an advection equation:

$$\frac{\partial \varphi}{\partial t} + \vec{v}_{fil} \cdot \nabla \varphi = 0$$

where $\vec{v}_{fil}$ represents the velocity of filament growth.
Stabilization of LSM

We adapt a stabilization scheme previously proposed by Olsson ’02.

\[ \frac{\partial \varphi}{\partial t} + \text{div}(\varphi(1 - \varphi)\vec{n}) - \xi \Delta \varphi = 0 \]

where \( \vec{n} \) represents the normal at the interface solution of:

\[ \vec{n} - \chi \Delta \vec{n} = \frac{\nabla \varphi}{\|\nabla \varphi\|} \]

with the boundary condition

\[
\begin{cases}
\vec{n} = 0 & \text{on } B_1 \cup B_3 \\
\vec{n} = (f(x), 0)^t & \text{on } B_2
\end{cases}
\]

- The boundary condition: to obtained the normal at the boundary with a good orientation.
- The diffusive term \( \chi \Delta \vec{n} \) is adjusted to regularize the normal near the level set \( \varphi = 0.5 \) (normal presents numerical instabilities).
We take

- $\xi = h$, where $h$ is the characteristic mesh size.
- $\chi = 10^{-17}$ results from numerical tests.

The level set equation is discretized with the scheme:

$$\frac{\varphi_{k+1}^n - \varphi_k^n}{\Delta \tau} + \text{div} \left( \frac{\varphi_{k+1}^n + \varphi_k^n}{2} - \varphi_{k+1}^n \varphi_k^n \right) = \xi \nabla \left( \frac{\varphi_{k+1}^n + \varphi_k^n}{2} \right) \cdot \vec{n} \text{div}(\vec{n})$$

**Figure**: Representation of $f$ in order to obtain a good boundary condition
**Figure:** Results of advection equation simulation with stabilization scheme and no clusters. (a) Initial Filament, (b) Filament at time $t = 150\,\text{ns}$, (c) $\varphi$ at time $t = 150\,\text{ns}$, (d) Filament at time $t = 260\,\text{ns}$, (e) $\varphi$ at time $t = 260\,\text{ns}$
**FIGURE:** Left: Sketch of an Ag/Ag-Ge-Se/Pt cell with nanodispersed Ag$_2$Se particles in a Ge-Se matrix. a) OFF state; b) ON state. Right: High Resolution TEM image of the Ag-doped GeSe-based solid electrolyte showing the presence of Ag-rich clusters.
**Figure**: Right: Filament growth with clusters in the electrolyte at $t = 1.45\mu s$; Left (a) Reinitialization of $\varphi$ after a nucleation step at $t = 1.1\mu s$. (b) Filament at $t = 1.15\mu s$. 
Clusters are used by electrons as bridge in the electrolyte when a filament touch a cluster, so the switching time decreases. The presence of clusters could explained the variability in switching time measurements.

**Figure:** Experimental and numerical switching time ($t_{SET}$) for CBRAM devices with a chalcogenide ($GeS_2$) electrolyte. Numerical switching time gives different results with clusters in the electrolyte.
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The model

**Hypothesis**: Constant velocity $v_{\Gamma_{fil}}$

\[
ed \frac{\partial C_{Ag}^+}{\partial t} - \text{div}(\epsilon D \nabla C_{Ag}^+ + \mu C_{Ag}^+ \nabla V) = 0
\]

\[
\begin{cases}
-(\epsilon D \nabla C_{Ag}^+ + \mu C_{Ag}^+ \nabla V).\vec{n} = J_{BV} & \text{on } \Gamma_+
\\
-(\epsilon D \nabla C_{Ag}^+ + \mu C_{Ag}^+ \nabla V).\vec{n} = J'_{BV} & \text{on } \Gamma_{fil}
\\
-(\epsilon D \nabla C_{Ag}^+ + \mu C_{Ag}^+ \nabla V).\vec{n} = 0 & \text{on } \Gamma_{ext}
\end{cases}
\]

\[-\text{div}(\epsilon \nabla V) = eC_{Ag}^+\]

\[
\begin{cases}
V = V_{app} & \text{on } \Gamma_+
\\
V = 0 & \text{on } \Gamma_{fil}
\\
\nabla V.\vec{n} = 0 & \text{on } \Gamma_{ext}
\end{cases}
\]
Solutions of the PDEs system

In the following to simply notations we denote \((C, V) = (C_{Ag^+}, V)\) and \(eD = D, e\mu = \mu\)

**Lemma**

For all \(t > 0\), \(C(t, x) > 0\) and \(C \in L^2(\Omega(t))\).

**Definition**

We say a function couple \((C, V) \in L^2(0, T; H^1(\Omega(t))) \times L^2(0, T; H^3(\Omega(t)))\) with \(\frac{\partial C}{\partial t} \in L^2(0, T; [H^1(\Omega(t))]')\) is a weak solution if for all \(w \in H^1(\Omega(t))\)

\[
\int_{\Omega(t)} \frac{\partial C}{\partial t} w + \int_{\Omega(t)} (D \nabla C + \mu C \nabla V) \nabla w - \int_{\Gamma_+} J_{BV} w + \int_{\Gamma_{fil}(t)} J'_{BV} w = 0
\]

\[
\int_{\Omega(t)} \nabla V \nabla w = \int_{\Omega(t)} C w
\]
Theoretical analysis

ALE method

We apply an ALE (Arbitrary Lagrangian Eulerian) method

- The main idea: transform the system from the moving domain $\Omega_t$ to a fixed reference domain $\hat{\Omega}$.
- We assume that, there exists a transformation:

$$\varphi : \hat{\Omega} \times [0, T] \rightarrow \bigcup_{t \in [0, T]} \Omega(t)$$

$$(\hat{x}, t) \rightarrow x_t = \varphi(\hat{x}, t),$$

where

- $\hat{x}$ are the points of the reference domain $\hat{\Omega}$
- $x_t$ are the points of the moving domain $\Omega(t)$.

- We define the domain velocity $\vec{v}_{\Gamma_{fil}}$ as:

$$\vec{v}_{\Gamma_{fil}}(\hat{x}, t) := \frac{\partial}{\partial t} \varphi(\hat{x}, t).$$
ALE method

- Under the transformation $x = \varphi(\hat{x}, t)$ we have
- $(C, V) \rightarrow (\hat{C}, \hat{V})$

$$\int_{\hat{\Omega}} \left( \frac{\partial \hat{C}}{\partial t} - \frac{\partial \varphi}{\partial t} \cdot M_J \nabla \hat{x} \hat{C} \right) \hat{w} \left| \det J_\varphi \right| + \int_{\hat{\Omega}} (DM_J \nabla \hat{C} + \mu \hat{C} M_J \nabla \hat{V}) \cdot M_J \nabla \hat{w} \left| \det J_\varphi \right| \right) \hat{w} \left| \det J_\varphi \right|$$

$$- \int_{\hat{\Gamma}_+} J_B V \hat{w} \left| \det J_\varphi \right| + \int_{\hat{\Gamma}_{fil}} J'_B V \hat{w} \left| \det J_\varphi \right| = 0$$

$$\begin{cases} \int_{\hat{\Omega}} M_J \nabla \hat{V} \cdot M_J \nabla \hat{w} \left| \det J_\varphi \right| - \int_{\hat{\Omega}} \hat{C} \hat{w} \left| \det J_\varphi \right| = 0 \\ \hat{V} = V_{app} \text{ sur } \hat{\Gamma}_+ \\ \hat{V} = 0 \text{ sur } \hat{\Gamma}_{fil} \end{cases}$$

- $J_\varphi(\hat{x}, t)$ represents the jacobian matrice of $\varphi$ in $(\hat{x}, t)$
- We assume there exits $\varpi, \vartheta > 0$ such as

$$\forall (\hat{x}, t) \in \hat{\Omega} \times [0, T], \quad \vartheta < \left| \det J_\varphi(\hat{x}, t) \right| < \varpi.$$
Galerkin approximation

Let \((\hat{w}_k)_{k \in \mathbb{N}^*}\) a function indexed family. We assume

- It is an orthogonal basis of \(H^1(\hat{\Omega})\)
- An orthonormal basis of \(L^2(\hat{\Omega})\).
- This base is used to approach \(\hat{C}\) using the Galerkin method.

We define the Galerkin approximation of \(\hat{C}\) with

\[
\hat{C}_m : [0, T] \rightarrow H^1(\Omega_t) \\
t \mapsto \hat{C}_m(t) = \sum_{k=1}^{m} d_k^m(t) \hat{w}_k
\]

The function \(\hat{C}_m\) is defined as a Galerkin approximation

\[
\forall k, 1 \leq k \leq m, \\
\int_{\hat{\Omega}} \left( \frac{\partial \hat{C}_m}{\partial t} - \frac{\partial \varphi}{\partial t} \cdot M_J \nabla \hat{x} \hat{C}_m \right) \hat{w}_k | \det J_\varphi | + \int_{\hat{\Omega}} (D M_J \nabla \hat{C}_m + \mu \hat{C}_m M_J \nabla \hat{v}(\hat{C}_m) \cdot M_J \nabla \hat{w}_k | \det J_\varphi | + \int_{\Gamma_{BV}} J'_B V \hat{w}_k | \det J_\varphi | = 0
\]
Galerkin approximation

We assume $\hat{V}(\hat{C}_m)$, depending on $\hat{C}_m$, is a solution (in $H^1(\hat{\Omega})$) of

$$
\begin{cases}
\int_{\Omega} M_J \nabla \hat{V} \cdot M_J \nabla \hat{w} \mid \det J_\varphi \mid - \int_{\Omega} \hat{C}\hat{w} \mid \det J_\varphi \mid = 0 \\
\hat{V} = V_{app} \text{ sur } \hat{\Gamma}_+ \\
\hat{V} = 0 \text{ sur } \hat{\Gamma}_{fil}
\end{cases}
$$

Lemma

For each integer $m \in \mathbb{N}^*$, there exists a unique couple $(\hat{C}_m, V(\hat{C}_m))$ satisfying the above equations on $[0,T_m]$

We denote

$$D_m(t) = (d^i_m(t))_{i \in [1,m]}^t$$

$$M = \left[\int_{\Omega} \hat{w}_i \hat{w}_j \mid \det J_\varphi \mid \right]_{(i,j) \in [1,m]^2}$$

we have

$$\frac{\partial}{\partial t} D_m(t) = M^{-1} F(D_m)$$
Energy estimates

- We need to show that a subsequence of our solutions \((\hat{C}_m, \hat{V}_m)\) of the approximate problem converges to a weak solution.
- We need uniform estimates

**Lemma**

*There exists \(T > 0\) independent of \(m\) and a constant \(C > 0\) such as for all \(m \geq 1\), \(T_m \geq T\) we have*

\[
\sup_{t \in [0,T]} \| \hat{C}_m \|_{L^2(\hat{\Omega})}^2 + \int_0^T \| \hat{C}_m \|_{H^1(\hat{\Omega})}^2 + \int_0^T \left\| \frac{\partial}{\partial t} \hat{C}_m \right\|_{[H^1(\hat{\Omega})]'}^2 \leq C
\]

\[
\int_{\hat{\Omega}} \hat{C}_m^2 \leq \int_{\Omega(t)} C_m^2
\]
Energy estimates

Proof idea

\[ \int_{\tilde{\Omega}} \left( \frac{\partial \hat{C}_m}{\partial t} - \frac{\partial \varphi}{\partial t} \cdot M_J \nabla \hat{C}_m \right) \hat{w}_k | \det J_\varphi | + \int_{\tilde{\Omega}} (D M_J \nabla \hat{C}_m + \mu \hat{C}_m M_J \nabla \hat{v}(\hat{C}_m) \cdot M_J \nabla \hat{w}_k) - \int_{\Gamma_+} J_{BV} \hat{w}_k | \det J_\varphi | + \int_{\Gamma_{fil}} J'_{BV} \hat{w}_k | \det J_\varphi | = 0 \]

\[ \int_{\Omega(t)} \frac{\partial C}{\partial t} w + \int_{\Omega(t)} (D \nabla C + \mu C \nabla V) \nabla w - \int_{\Gamma_+} J_{BV} w + \int_{\Gamma_{fil}(t)} J'_{BV} w = 0 \]

\[ \int_{\hat{\Omega}} \hat{u}_m^2 \leq \vartheta \int_{\hat{\Omega}} \hat{u}_m^2 | \det J_\varphi | \leq \int_{\Omega(t)} u_m^2 \]
Regularity of Poisson equation with moving boundaries

In this part, we want to prove the following result

**Theorem**

Let us take $\Omega(t)$ a $C^3$ open set with $\Gamma$ bounded. Let $V^t \in H^1(\Omega(t))$ solution of

\[
\begin{align*}
-\Delta V^t &= C^t \\
V^t &= V_{app \text{ sur } \Gamma_+} \\
V &= 0 \text{ sur } \Gamma_{fil}
\end{align*}
\]

with $C^t \in H^1(\Omega(t))$, then $\forall t \in [0,T]$ $V^t \in H^2(\Omega(t)) \cap H^3(\Omega(t))$ and there exist a constant $K > 0$ which is not time dependent such as for all $t \in [0,T]$,

\[
\|V(C^t)\|_{H^2(\Omega(t))} \leq K(\|C^t\|_{L^2(\Omega(t))} + \|V_{app}\|_{H^{3/2}(\Gamma_+)}) \quad \text{and} \\
\|V(C^t)\|_{H^3(\Omega(t))} \leq K(\|C^t\|_{H^1(\Omega(t))} + \|V_{app}\|_{H^{5/2}(\Gamma_+)})
\]
Regularity of Poisson equation with moving boundaries
Existence and unicity of solutions

**Theorem**

There exists an unique solution

\[(C, V) \in L^\infty(0, T; L^2(\Omega_t)) \cup L^2(0, T; H^1(\Omega(t))) \times L^2(0, T; H^3(\Omega(t)))\]

Take the limit \(m \to +\infty\).

So, it exists a subsequence \((\hat{C}_{m_l})_{l \in \mathbb{N}^*}\) of \((\hat{C}_m)_{m \in \mathbb{N}^*}\) and a subsequence \((\hat{V}_{n_k})_{k \in \mathbb{N}^*}\) of \((\hat{V}_n)_{n \in \mathbb{N}^*}\) such as

\[
\begin{align*}
\hat{C}_{m_l} &\rightharpoonup \hat{C} \text{ weak in } L^2\left(0, T; H^1(\hat{\Omega})\right) \\
\hat{C}_{m_l} &\to \hat{C} \text{ strong in } L^2\left(0, T; L^2(\hat{\Omega})\right) \\
\hat{C}_{m_l}' &\rightharpoonup \hat{C}' \text{ weak in } L^2\left(0, T; H^{-1}(\hat{\Omega})\right) \\
\hat{V}(\hat{C}_{m_l}) &\rightharpoonup \hat{V} \text{ weak in } L^2\left(0, T; H^2(\hat{\Omega})\right)
\end{align*}
\]
Figure: Results of simulation with constant velocity.
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Work in progress and perspectives

Analysis of existence and unicity of solutions with speed $v_{fil}$ depending on $C$.

- $v_{fil} = \frac{V_{Ag}}{e} J_{BV} \vec{n}$, ($\vec{n}$ the exterior normal ar $\Gamma_{fil}$)
- $J_{BV} = \text{const} \sqrt{C}$

$$\int_{\Omega(t)} \frac{\partial C}{\partial t} w + \int_{\Omega(t)} (D \nabla u + \mu C \nabla V) \nabla w - \int_{\Gamma_+} J_{BV} w + \int_{\Gamma_{fil}(t)} J'_{BV} w = 0$$

$$J_{BV} \rightarrow (C, V) \rightarrow \sqrt{C}|_\Gamma = \tilde{J}_{BV}$$

- We do not have compatibility between the regularity of $C$ and the regularity of $\Omega$. 
Work in progress and perspectives

Add tunneling effect into the model
Conclusions and perspectives

- **Introduction of a stochastic model** (with J. Garnier, F. Hecht, and Y. Maday)

**Figure**: Sketch of an Ag/Ag-Ge-Se/Pt cell with nanodispersed $Ag_2Se$ particles in a Ge-Se matrix.
Conclusions and perspectives

- Mathematical modeling for OXRAMs memories

**Figure:** OXRAM memory schematic view
Merci !