# Coupling the Phase-Field Method with an Electrothermal Solver to Simulate Phase Change Mechanisms in PCRAM Cells

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Abstract—In order to simulate the electro-thermal characteristics of our devices and to reproduce the phase change mechanisms of the PCM material during the set and reset operations, we have developped a finite element electro-thermal solver coupled with the Phase Field Method. Retention simulations for full GST layers are first presented. Then set simulations, with a low and with a high current, starting from a partially amorphized domain are presented. The different cristallized domains for low and high set current highlighted by TEM images reveal the impact of the set current on the crystallization mechanisms. The Phase-Field simulations confirm this result.

# I. INTRODUCTION

The Phase Field Method (PFM) has become a powerfull tool for simulating the microstructural evolution at the mesocale in a wide variety of material processes, such as solidification, solid-state phase transformations, precipitate growth and coarsening [1]. An important advantage of the phase field method is that there is no need to follow explicitly the position of the interfaces during the microstructural evolution. Phase-Change Memories (PCM) are today considered the most mature among novel non-volatile memory technologies. Thanks to an optimized Ge-rich GeSbTe alloy, we have demonstrated a good trade-off between set speed and data retention performances [2]. In order to simulate the electrothermal characteristics and to reproduce the phase change mechanisms during set and reset operations, we have developped a finite element electro-thermal solver coupled with the PFM. An explicit treatment of nucleation was coupled to the PFM. The PFM parameters of our simulator were adjusted from global experimental results (retention time, crystallization kinetic during set operation). Retention simulations are first presented to illustrate the capability of our simulator to represent stochastic nucleation followed by the growth of critical nuclei. Simulations of set operations with low and high current are then presented and analyzed. The PFM simulations enlighten the crystallization dynamics at the origin of the peculiar microstructure observed experimentally in set devices programmed with different currents.

# II. THE PHASE-FIELD METHOD APPLIED TO THE PHASE-CHANGE IN PCM

The PFM is considered in this work in a phenomenological approach and the phase field  $\eta$  is used to distinguish two

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coexisting phases.  $\eta$  is a non conserved order parameter representing the local crystallinity of PCM material:  $\eta$ =0 in the disordered phase (liquid/amorphous) and  $\eta$ =1 in the ordered phase (crystalline). It evolves continuously from its value in the disordered phase to its value in the ordered phase.

#### A. Equations of the Phase-Field Method

Time evolution of  $\eta$  is governed by the Allen-Cahn equation (1) and corresponds to the reduction of the total energy of the system composed of free-energy of bulk phases and energy of interfaces between phases [3].

$$\frac{\partial \eta}{\partial t} = -L_{\eta} \left( \frac{\partial f(\eta)}{\partial \eta} - \kappa \nabla^2 \eta \right) \tag{1}$$

where  $L_{\eta}$  is a positive kinetic coefficient,  $f(\eta, T)$  is a local free-energy density that consists of a double well function  $f_{dw}$  and an interpolation function  $f_p(\eta, T)$ .

$$f(\eta, T) = f_{dw}(\eta) + f_p(\eta, T)$$
(2)

The double well potential  $f_{dw}(\eta) = H\eta^2(1-\eta)^2$  has minina at 0 and 1. H is the depth of the energy wells.

The interpolation function  $f_p(\eta, T)$  combines the free energy expressions of the amorphous and crystalline phases.

$$f_p(\eta, T) = g(\eta)G^{\rm am}(T) + (1 - g(\eta))G^c(T)$$
(3)

where g must verify: g(0)=1,g(1)=0,g'(0)=0,g'(1)=0. For g, we have retained the expression used in [4].

$$g(\eta) = (1 - \eta)^3 (-5 + 15\eta + 6(1 - \eta)^2)$$
(4)

Finally, as  $\Delta G(T) = G^{(am)}(T) - G^{c}(T)$ , the free-energy density can be written:

$$f(\eta, T) = f_{dw}(\eta) + g(\eta)\Delta G(T)$$
(5)

At the melting point, the two phases have the same free energy density ( $\Delta G = 0$ ) and  $f(\eta, T_m)$  reduces to the symmetric double well potential  $f_{dw}(\eta)$ . At the melting point, there is no driving force for cristallization but the two phases are separated by a free energy barrier and, phenomenologically, this is captured by the double-well potential.

The kinetic coefficient  $L_{\eta}$ , introduced in Eq. (1), can be calculated from the front velocity (which in our case is the crystalline growth velocity  $V_{q}$ ) by the equality:

$$L_{\eta} = \frac{V_g \cdot R_c}{\kappa} \tag{6}$$

where  $R_c$  is the curvature radius of the interface that we will consider as equal to the radius of critical nuclei [3] [4]. Main thermodynamical parameters are given in Table I. The

Description	Symbol (unit) Value	
Melting temperature	$T_m(K)$	889
Glass temperature	$T_g(K)$	353
Latent Heat of melting	$L(J/m^3)$	$1.1 \cdot 10^9$
Bulk free energy difference	$\Delta G(J/m^3)$	$\frac{L(T_m - T)}{T_m}$
Interfacial energy	$\sigma_a(J/m^2)$	0.2
Nucleation barrier	$\Delta G^*(J)$	$\frac{4\sigma_a^3}{27\Delta G^2}$
Stationary Nucleation rate	$I_s(s^{-1}.m^{-3})$	$C_N exp(-\frac{\Delta G^*}{kT})$
Inter-atomic distance	d(m)	$3 \cdot 10^{-10}$
Growth Velocity	$V_g(m/s)$	$d\mu \left(1 - exp(\frac{\Delta G d^3}{kT})\right)$

TABLE I. MAIN THERMODYNAMICAL PARAMETERS FOR GST

parameter  $\mu$  is a function of temperature and represents the frequency of accretion of an atom to a crystalline nuclei.  $C_N$  is a pre-factor coefficient for the nucleation rate.

# B. Choice of the PFM parameters

The parameters of the PFM method,  $\kappa$  and H, are considered constant for this work. In addition to this, two other significative parameters are introduced:  $\gamma$  and  $d_{\text{int.}}$   $\gamma$  is the excess free energy due to the presence of the energy wall considered between the two phases at equilibrium. It can be evaluated analytically from  $\kappa$  and H [1]:

$$\gamma = \frac{\sqrt{\kappa H}}{3\sqrt{2}} \tag{7}$$

It has the unit of an interfacial energy  $(J/m^2)$ .

The parameter  $d_{\rm int}$  can also be evaluated analytically from  $\kappa$  and H:

$$d_{\rm int} = \sqrt{\frac{\kappa}{H}} \tag{8}$$

It has the unit of a length and represents the interface thickness. Parameters  $\kappa$  and H are not directly extracted from material properties but are chosen to have  $\gamma$  in the range commonly accepted for interfacial energies for our material and to obtain a kinetic of crystallization with PFM simulations that fits with kinetic of crystallization extracted from experimental results.

TABLE II. PHASE-FIELD MAIN AND RELATED PARAMETERS

Description	Symbol	Value
Gradient energy coefficient	κ	$3.84 \cdot 10^{-11} J/m = 2.5 eV/nm$
Potential well height	Н	$0.32 \cdot 10^9 J/m^3 = 2eV/nm^3$
Interfacial energy	$\gamma$	$\frac{\sqrt{\kappa H}}{3\sqrt{2}} = 0.08J/m^2$
Interfacial thickness	d	$\sqrt{\frac{\kappa}{H}} = 0.55 \cdot 10^{-9} m$
Curvature radius	$R_c(m)$	$\frac{2\gamma}{\Delta G}$

### III. NUCLEATION

Phase change mechanisms that occur during the PCM operations generally involve concurrent nucleation and growth. Nucleation could be introduced in the PFM as an additional Langevin noise term in Eq. (1) but this can become computationnaly expensive since it requires sampling at a very high frequency if nucleation events are very rare. In our work as proposed by [5], nucleation model will be maintained as a separate entity from the Phase-Field equation. Our approach is to have two algorithms which alternate, one for nucleation and one for growth and coarsening corresponding to the advance of Eq. (1).

As already presented [6], our nucleation model relies on the classical nucleation theory. Due to the very fast increase (in the order of  $10^{12}K/s$ ) and decrease of temperature (in the order of  $10^{10}K/s$ ) associated to the electrical pulses used in PCM, a transient nucleation rate is taken into account [7]:

$$I(x, y, t) = I_s(x, y) \cdot exp(-\frac{\tau}{t})$$
(9)

where  $I_s$  is the stationary nucleation rate and  $\tau$ , the time needed to establish the stationary state population of nuclei. In practice, in our time algorithm, the term  $exp(-\frac{\tau}{t})$  is evaluated using  $\delta t$  the isothermal holding time instead of t;  $\delta t$  depends on the temperature variation rate. The expected number of critical nuclei generated during dt in an unit volume  $h^3$  is calculated by:

$$N(x, y, t) = I(x, y, t) \cdot dt \cdot h^3 \tag{10}$$

During the nucleation step, critical nuclei are introduced into individual cells randomly but with a mean formulation rate given by N(x, y, t).

Nucleation is explicitly introduced in the PFM algorithm using discretization of time derivative in equation (1)

$$\frac{\partial \eta}{\partial t} = \frac{\eta^n - \eta^*}{dt^n} \tag{11}$$

where  $\eta^*$  is the update of  $\eta$  to take into account new critical nuclei and  $\eta^n$  represents the variable  $\eta$  at given time  $t^n$  in the discretized time.

$$\eta^{*}(x, y, t^{n+1}) = \begin{cases} \eta(x, y, t^{n}) & \text{if } r(x, y, t^{n}) \ge N(x, y, t^{n}) \\ 1 & \text{if } r(x, y, t^{n}) < N(x, y, t^{n}) \end{cases}$$
(12)

where r(x, y, t) is a random number following a discrete uniform distribution between 0 and 1.

# IV. EQUATIONS OF THE ELECTRO-THERMAL SOLVER

An ohmic model approach is used to simulate the electrical behavior of the PCM cell. The electro-thermal solver relies on the coupled system of partial differential equations formed by the current conservation equation and the heat transfer equation.

$$\nabla \cdot (-\sigma \nabla V) = 0 \tag{13}$$

$$\rho C_p \frac{\partial T}{\partial t} + \nabla \cdot (-k_{th} \nabla T) = \sigma (\nabla V)^2$$
(14)

where  $\sigma$ ,  $\rho$ ,  $C_p$  and  $k_{th}$  stand for the materials electrical conductivity, density, heat capacity and thermal conductivity. Electrical and thermal conductivity of the phase change material are phase dependent. The electrical conductivity in the amorphous phase is modelled by an off-state if the local

electric field is lower than a threshold electric field  $(E_{th})$  and switches to an on-state if the local electric field becomes higher than  $E_{th}$  [8]. Once the material has locally switched to the on-state, our model includes a possible recovery of the offstate if the local density of current gets lower than a holding current density  $(j_{hold})$ . We consider that  $E_{th}$  and  $j_{hold}$  are parameters to be adjusted. For Ge-rich GST we have retained:  $E_{th} = 2.975 \cdot 10^6 [V/m]$  and  $j_{hold} = 10^9 [A/m^2]$ 

$$\sigma_{\rm on} = \frac{\sigma_0}{2} \left( \tanh(B_e T + C_e) + D_e \right) \tag{15}$$

$$\sigma_{\text{off}} = \frac{A}{\|\nabla V\|} \exp(-\frac{E_{am}}{k_B T}) \sinh(\frac{qB \|\nabla V\|}{k_B T}) \qquad (16)$$

The thermal conductivity of amorphous phase in the electricaloff state is considered constant while the thermal conductivity of the phase change material in the electrical-on state  $(k_{th}^{on})$ depends on the temperature in a similar way than  $\sigma_{on}$ .

$$k_{th}^{\rm on} = \frac{k_{th}^0}{2} \left( \tanh(B_{th}T + C_{th}) + D_{\rm th} \right)$$
(17)

A set of parameters for electrical and thermal conductivity for Ge-rich GST and for the heater was fixed conjointly in order to obtain current-voltage curves close to experimental results.

## V. IMPLEMENTATION

The system of previous equations is discretized using the Finite-Element method with the Partial Derivative Equation interfaces of COMSOL. In PCM material, electrical and thermal conductivities depend on a State variable that is locally crystalline, liquid, amorphous off or amorphous on. Variable State is updated from the variable  $\eta$  and the electrical state (on-state or off-state). This leads to a strongly coupled and non linear set of equations which is solved self-consistently using a non linear Newton solver.

#### VI. SIMULATION RESULTS AND DISCUSSION

#### A. Retention simulations

In order to validate the ability of our model to reproduce nucleation and growth in simplified thermal conditions first, retention simulations are carried out in a layer of fully amorphous  $Ge_2Sb_2Te_5$ . We simulate the crystallization of  $Ge_2Sb_2Te_5$  as a first application because it crystallizes with no change in composition. Eq. (1) is coupled to the heat equation in which the exothermic heat associated to crystallization is taken into account.

$$\rho C_p \frac{\partial T}{\partial t} - L \frac{dh}{d\eta} \frac{\partial \eta}{\partial t} = \nabla \cdot \left( -k_{th} \nabla T \right)$$
(18)

where  $h(\eta)$  is an smooth interpolation function verifying h(0)=1,h(1)=0,h'(0) and h'(1)=0; we have chosen h equal to g.

For these simulations a ramp of temperature with a slope of 10K/min is applied as a boundary condition to the layer. Images extracted from the simulations (Fig. 1) at different temperatures illustrate the emergence of critical nuclei and their growth. The resistance of the partially cristallized layer is calculated with our electro-thermal model based on a phase dependent conductivity. As illustrated by (Fig. 2) the simulated



Fig. 1. Evolution of  $\eta$  during the retention simulation

crystallization temperature is around 160°C and it is in agreement with the crystallization temperature commonly accepted for  $Ge_2Sb_2Te_5$ .



Fig. 2. Resistance versus temperature during the retention simulation

#### B. Low and High current simulations

The devices simulated are state-of-the-art PCM devices [9] comprising wall storage elements (Fig. 3) on an aggressive 90nm CMOS technology platform. The width of the devices is 180nm. The phase change material encapsulated in these



Fig. 3. Schematic drawing of the PCM Wall storage element

devices is an optimized alloy Ge-rich GST. Experimental results presented in [10] show that the crystallization temperature (Tc) of this alloy is about 200°C higher than that of  $Ge_2Sb_2Te_5$ . To take into account the increase of Tc in our crystallization model, we use a reduced nucleation rate and a reduced crystalline growth velocity compared to that of  $Ge_2Sb_2Te_5$ . Starting from the amorphous dome obtained by the simulated reset state, set simulations with low current ( $600\mu A$ ) and high current ( $1000\mu A$ ) are realized. The programming pulses used correspond to a plateau of 500ns and a trailing edge of 500ns (resp. 1000ns) for low current pulse (resp. high current pulse). The simulations qualitatively reproduce the formation of a conductive filament where the material has switched from the off state to the on state. Then the vicinity of the conductive filament is heated up leading to a melted domain inside the initial amorphous dome. The size of the melted domain depends on the intensity of the set current (Fig. 4). We observe that, during the set operation with low and



Fig. 4. Comparison of the melted area for set with low and high current

with high current, nucleation has no role in the crystallization which is only due to the regrowth of the crystalline/amorphous interface. The isovalue curves for growth velocity coincide with the isothermal curves in the amorphous phase what means that the growth velocity is determined by the shape of the isothermal curves. With the low current set, the temperature field leads to the growth of the top crystal material toward the inside of the amorphous dome. At the end of the set low operation, two amorphous domains remain at the left and at the right of the recrystallized column. (Fig. 5).



Fig. 5. Simulation of the phase change during the low current SET pulse

With the high current, the melted region starts to get uniformly smaller after 500ns until it dissapears at 800ns leaving the whole cell in crystalline phase (Fig. 6). The



Fig. 6. Simulation of the phase change during the high current SET pulse

simulations confirm the results extracted from TEM images (Fig. 7) and illustrate how in the low current set a single crystalline column of single orientation is formed because the crystallization proceeds by growth of a pre-existing grain. On the contrary with the high current set a polycrystalline structure is obtained [10].



Fig. 7. Extracted from [10] TEM BF/DF images for the set states of Ge-rich GST devices a) Operating the set at low current, a single crystalline column of single orientation is formed across the active area b) Operating the set at high current results in a polycrystalline structure with randomly oriented grains

#### VII. CONCLUSION

We have developped a finite-element electro-thermal solver coupled with the Phase-Field Method. Simulations of reset and set operations with electronic switching for state-of-theart PCM devices have been realized. Our simulations give important insights into the influence of set current on the shape of the crystallized domain. Our phase change model has been developped at first for the reference alloy  $Ge_2Sb_2Te_5$  and corresponds to crystallization without composition change. We will go further by modeling the phase change of the Ge-rich GST alloy that proceeds with composition change and phase separation. The crystallization dynamic will be more accurately simulated taking into account nucleation and growth separately for Ge and GST phases. The Multi Phase-Field formulation could be a good method for that purpose.

#### ACKNOWLEDGMENT

The authors would like to thank Patrick Namy from SIMTEC for fruitful discussions about implementation of our model in comsol.

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