Optimal Control of Mean Field Models for Phase Transitions

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Abstract: Various models prescribe precipitation due to phase transitions. On a macroscopic level the well-known Lifshitz-Slyozov-Wagner (LSW) models and its discrete analogons, so-called mean field models, prescribe the size evolution of precipitates for two-phase systems. For industrial tasks it is desirable to control the resulting distribution of droplet volume. While there are optimal control results for phase-field models and for nonlinear hyperbolic conservation laws, it seems that control problems for LSW equations and mean field models, including measure-valued solutions or switching conditions, have not been considered so far. We formulate the model for this important new control problem and present first numerical results.

Keywords: Optimal control, Process control, Nonlinear control systems, End point control, Partial differential equations, Characteristic curves, Differential algebraic equations (DAE), Mathematical models, Phase transition, LSW equation

1. MODELS FOR PHASE TRANSITIONS

Phase transitions are an important phenomenon in material science. On the one hand phase transitions may be exploited in order to design a requested material, on the other hand they may destroy desirable properties of designed materials. For example, the industrial production process of semi-insulating gallium arsenide (GaAs) requires at the end some additional heat treatment at high temperatures in order to ameliorate the quality of the semi-insulator.

One of the challenges is the necessity to guarantee a mean mole fraction of arsenic (As) in the GaAs wafer of $X_0 = 0.500082$ within high accuracy, in order to have the desired semi-insulating behaviour. During this final heating process unwanted liquid droplets precipitate in the solid crystal due to misfits and due to supersaturation. These precipitates influence negatively mechanical and semi-insulating properties of the crystal. Their elimination, if possible, is a crucial point for the production of semi-insulators.

For the modelling of phase transitions various types of models have been suggested. Sharp-interface models and phase-field models, where the interface is smeared out in the latter, capture the spatial structure of a phase transition, while macroscopic models, like the LSW model (Lifshitz and Slyozov (1961); Wagner (1961)) or the mean field model, and BD models (Becker and Döring (1935)) do not. Sharp-interface models, phase-field models and macroscopic models are continuous diffusion models while the BD model is an atomic nucleation model. Macroscopic models may be justified rigorously as homogenization limits of sharp-interface models or of BD models for small droplet volume fraction.

We consider a mathematical model that describes the evolution of the precipitates including surface tension and bulk stresses on a macroscopic scale. It is obtained by homogenization of a sharp-interface model, derived from thermodynamical principles in Kimmerle (2009). We examine the corresponding control problem. While results exist for the optimal control of phase-field models, e.g. for the Allen-Cahn equation (Farshbaf-Shaker (2011a,b)) or Cahn-Hilliard equation (Hintermüller and Wegner (2011)), the control of a macroscopic model has not been considered so far as known by the author. Instead of the well-established LSW model our model comprises the microstructure of the crystal within the diffusion process, mechanical deformations within linear elasticity, and the fact that droplets with only a few atoms do not behave like a liquid. The latter is modelled by the introduction of a minimal droplet volume $V_{min} > 0$. Our model is a realistic model for phase transitions between liquid and solid including linear elasticity and is not only restricted in its applicability to semi-insulating GaAs.

We focus on the homogeneous version of this generalized LSW model, corresponding to the dilute scaling of droplet volume fraction. In the homogeneous LSW model the bulk is in quasi-static diffusion equilibrium. Different regimes for the motion of free boundaries, as volume-diffusion-controlled or interface-reaction-controlled interface motion can be considered. We assume homogeneous precipitates, i.e. the mass density and concentrations within the precipitates are constant.

2. CONTROL PROBLEM

We look for regimes, where for large times either only a few droplets, as small as possible, survive or where a homogeneous distribution $\nu_t(V)$ of the droplet volume V

may be achieved at a given final time t_f. This motivates the cost function

$$J(u_1,\nu_{t_f}) = \|u_1\|^2_{L^2(t_0,t_f)} + \int\limits_{V_{min}}^{\infty} (\alpha_1+\alpha_2 V)\, d\nu_{t_f}(V) \\ +\alpha_3 \int\limits_{V_{min}}^{\infty} \left| V \int\limits_{V_{min}}^{\infty} d\nu_{t_f}(S) - \int\limits_{V_{min}}^{\infty} S d\nu_{t_f}(S) \right|^2 d\nu_{t_f}(V) \tag{1}$$

where the positive weights α_k, $1 \le k \le 3$, may be chosen in principle as needed within the application. Note, that the integral terms are evaluated at the end point t_f and only the control cost depends on the whole time interval (t_0, t_f). A natural parameter control is provided by physical quantities like temperature u_1 or pressure. An initial control is provided by the total mass M and the total arsenic $N_1 = MX_0/m_1$, m_1 the molar mass of As, that are conserved, and, in principle, by the initial volume distribution of droplets $\nu_0(V)$. Since the pressure yields only a slight correction (Kimmerle (2009)), and it is not clear whether it is technically possible to influence precisely the initial distribution of droplets, we focus on the total mass $u_0 = M$ and the temperature u_1 as control.

2.1 Volume-diffusion-controlled regime

While droplets are parametrized by V, the bulk is parametrized by a fictive volume $\overline{V}$ corresponding to the mean field of the chemical potential. The evolution of the time-dependent non-negative measure $\nu_t(V)$, the density of droplet volume, is prescribed by the LSW equation

$$\partial_t \nu_t + a(\overline{V}, V, u_1)\, \partial_V \nu_t = 0 \text{ in } (V_{min}, \infty), \text{ a.e. in } [t_0, t_f], \tag{2}$$

while droplets smaller than V_{min} do not exist, yielding

$$\nu_t(V) = 0 \text{ in } [0, V_{min}], \text{ in } [t_0, t_f]. \tag{3}$$

The term a is the so-called Stefan condition, modelling the balance of mass/substance at the interface, and reads for the volume-diffusion-controlled regime

$$a(\overline{V}, V, u_1) = V^{1/3} \frac{\mu_I(\overline{V}, u_1) - \mu_I(V, u_1)}{\mathbb{X}(V, u_1)} \tag{4}$$

with a strictly positive function $\mathbb{X}$, monotone decreasing in V. Here $\mu_I(V, u_1)$ is the chemical potential of a precipitate, strictly monotone decreasing in the volume, with parameter u_1. From a it turns out that the chemical potential $\overline{\mu} = \mu_I(\overline{V}, u_1)$ is associated to the fictive volume $\overline{V}$ and droplets with chemical potential $\mu_I(V_i, u_1)$ smaller as $\overline{\mu}$ grow while droplets with a larger chemical potential shrink. The function

$$\mathbb{X}(V,u) = \frac{u_1}{B^D} \left[\frac{\rho_S(\overline{V},u_1)}{\eta_S(\overline{V},u_1)} \eta_L(V,u_1) - \rho_L(V,u_1) \right. \\ \left. + \left(\frac{\rho_S(\overline{V},u_1)}{\eta_S(\overline{V},u_1)} \partial_V \eta_L(V,u_1) - \partial_V \rho_L(V,u_1) \right) V \right] \tag{5}$$

results from the continuity of the flux of mass and substance over the interface. Note that the mass densities/concentrations ρ_L/η_L are evaluated on the liquid side of the interface, while ρ_S and η_S are evaluated on the solid side. B^D is a positive constant related to the mobility in the volume-diffusion-controlled regime.

The corresponding initial condition is

$$\nu(t_0, V) = \nu_0(V). \tag{6}$$

This is coupled to the conservation of mass/substance yielding an algebraic equation for $\overline{V}$ by means of the implicit function theorem,

$$\overline{V} = \zeta \left(\frac{M - \int_{V_{min}}^{\infty} \rho_L(V,u_1)\, V d\nu_t(V)}{MX_0 - m_1 \int_{V_{min}}^{\infty} \eta_L(V,u_1)\, V d\nu_t(V)}, u_1 \right) \\ \text{in } [t_0, t_f], \tag{7}$$

with a nonlinear, strictly monotone function

$$\zeta(\cdot, u_1) = \left(\frac{\eta_S(\overline{V}, u_1)}{\rho_S(\overline{V}, u_1)} \right)^{-1}. \tag{8}$$

The index of the algebraic equation (7) is 1.

Let $\mathcal{C} := C_0^0(0,\infty)$ and $\mathcal{C}'$ denote its dual space. Our control problem is to find states

$$\{\nu_t, \overline{V}\} \in C^0_{weak}([t_0,t_f], \mathcal{C}') \times C^0([t_0,t_f], \mathbf{R}), \tag{9}$$

an initial control parameter

$$u_0 \in \mathbf{R}^+, \tag{10}$$

and a control

$$u_1 \in L^\infty([t_0,t_f], \mathbf{R}^+), \tag{11}$$

s.t. the cost functional J, given in (1), is minimized under respect of the initial value problem for our DAE system (2) – (8), the pure state constraints,

$$\begin{array}{ll} a)\ \nu_t(V) \ge 0 & \forall V \in \mathbf{R}_0^+ \, \forall t \in [t_0,t_f], \\ b)\ \overline{V} \ge 0 & \forall t \in [t_0,t_f], \end{array} \tag{12}$$

and box constraints for the controls

$$\begin{array}{ll} u_{min,0} \le u_0 \le u_{max,0}, & \\ u_{min,1} \le u_1(t) \le u_{max,1} & \forall t \in [t_0,t_f], \end{array} \tag{13}$$

where $u_{min,j}$, $u_{max,j}$, $j = 0, 1$, are given strictly positive bounds.

Eq. (3) together with (4) implies that we switch from an ODE to the equation $\nu_t(V) = 0$ once a shrinking droplet reaches V_{min}.

2.2 Interface-reaction-controlled regime

In case of the interface-reaction-controlled regime we have again (2), (3), (6) – (13). We replace (4) by

$$a(\overline{V}, V, u_1) = V^{4/3} \frac{\mu_I(\overline{V}, u_1) - \mu_I(V, u_1)}{\mathbb{Z}(V, \overline{V}, u_1)} \tag{14}$$

where the balance of mass and substance at the interface between solid and a liquid droplet is encoded in the strictly positive function

$$\mathbb{Z}(V,\overline{V},u_1) = \frac{u_1}{B^I} \rho_L(\overline{V},V,u_1)^{1/2} \left[(\tilde{\mu}-1) \right. \\ - \left((\tilde{\mu}-1) \frac{\partial_V \rho_L(\overline{V},V,u_1)}{\rho_L(\overline{V},V,u_1)} + \tilde{\mu} \frac{\partial_V \eta_L(\overline{V},V,u_1)}{\eta_L(\overline{V},V,u_1)} \right. \\ \left. \left. - \frac{\partial_V \left(\rho_L(\overline{V},V,u_1) - m_1 \eta_L(\overline{V},V,u_1) \right)}{\rho_L(\overline{V},V,u_1) - m_1 \eta_L(\overline{V},V,u_1)} \right) V \right], \tag{15}$$

that is monotone decreasing in $\overline{V}$. $B^I > 0$ is a constant linked to the mobility in this regime and $\tilde{\mu}$ is the quotient of the molar mass of gallium and arsenide.

3. NUMERICAL SOLUTION

3.1 Mean field model

Numerically, we solve our problem by the method of lines. We consider a special case of our control problem, the so-called *mean field model*, where we assume a special initial condition

$$\nu_0(V) = \frac{1}{\mathcal{N}_0} \sum_{i=1}^{\mathcal{N}_0} \delta_{V_i^0}(V), \tag{16}$$

i.e. we have initially a discrete finite number $\mathcal{N}_0$ of distinct volumes

$$V_i(t_0) = V_i^0 \quad \forall i \in \{1; ...; \mathcal{N}_0\}. \tag{17}$$

The discrete mean field model amounts to solving our original LSW equation for a finite number $\mathcal{N}_0$ of characteristics.

We introduce another unknown $\mathcal{N}(t)$ encoding the number of droplets at time t with $V > V_{min}$. Precipitates below V_{min} vanish. We introduce t_j as the first time when $V_j \leq V_{min}$. If a precipitate never disappears, i.e. $V_j > V_{min}$ for all times, then we set $t_j = \infty$. For ease of notation we assume w.l.o.g. $V_1 \geq V_2 \geq .. \geq V_{\mathcal{N}_0 - 1} > V_{\mathcal{N}_0}$, thus it turns out that $V_{\mathcal{N}_0}$ vanishes first and V_1 remains as last droplet.

In this situation our control problem simplifies for both regimes to the following evolution of precipitates

$$\begin{array}{ll} \partial_t V_i & = a(\overline{V}, V_i, u) \text{ in } [t_0, t_f] \setminus \cup_{1 \leq j \leq \mathcal{N}_0} \{t_j\}, \\ V_i(t+) = V_i(t-) & \text{in } (\cup_{1 \leq j \leq \mathcal{N}_0} \{t_j\}) \cap [t_0, t_f], \end{array} \tag{18}$$

for $V_i > V_{min}$, while

$$V_i = 0 \text{ in } [t_0, t_f], \tag{19}$$

for $V_i \leq V_{min}$. Here we keep record of droplets below V_{min}, contrary to our original model, in order not to change the number of states with time what turns out to be more suitable for numerics. The ODE system is completed by the initial condition (17) and the conservation of mass/substance

$$\overline{V} = \zeta \left(\frac{M - \frac{1}{\mathcal{N}_0} \sum_{i=1}^{\mathcal{N}(t)} \rho_L(V_i, u_1)\, V_i}{MX_0 - m_1 \frac{1}{\mathcal{N}_0} \sum_{i=1}^{\mathcal{N}(t)} \eta_L(V_i, u_1)\, V_i}, u_1 \right) \quad \text{in } [t_0, t_f], \tag{20}$$

the so-called *mean field formula*. The state constraints read

$$\{V_i\}_{1 \leq i \leq \mathcal{N}}, \overline{V} \geq 0 \tag{21}$$

and we have the constraints (13) for the controls.

The system (17) - (21), (13) is called *mean field model*. Under reasonable assumptions on the data $\mathbb{X}$ or $\mathbb{Z}$, and $\mu_I, \zeta, \rho_L, \eta_L$, $u_{min,\cdot}$, and $u_{max,\cdot}$ we may solve the resulting discretized control problem with jumps in the states and its derivatives, both occurring whenever a droplet disappears. The numerical results rely on data for GaAs,

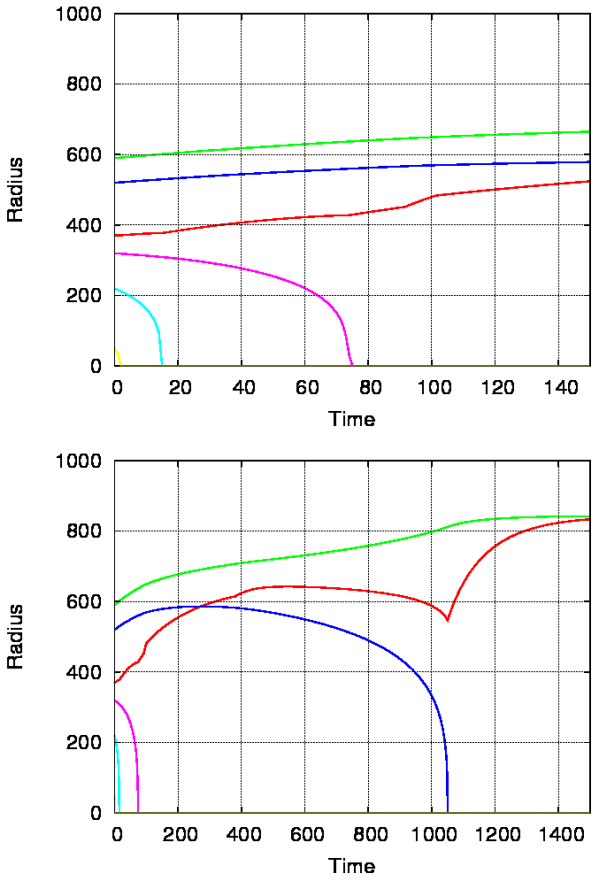

Fig. 1. Evolution of 5 droplet radii with initial radii 50 (yellow), 220 (cyan), 320 (magenta), 520 (blue), 590 (green), together with the bulk mean field radius (red) [10^{-9} m] vs. time [1 s]. Upper figure: short-time behaviour (up to 150 s), lower figure: long-time behaviour (up to 1500 s).

summarized in Dreyer and Kimmerle (2009). Note that the main influence of the control enters within (5) or (15). We study in the following the numerical solution in case of the volume-diffusion-controlled regime.

3.2 Numerical methods

We follow a direct method, meaning optimization of the discretized control problem corresponding to (17) – (21) and (13). For the update of the states we use central differences. Our optimization algorithm follows a sensitivity-based approach. This has been implemented in OCODE 1.5, a software code developed by Gerdts (2010).

Eq. (20) has index 1 and if we suppose a suitable initial condition

$$\overline{V}(t = t_0) = \overline{V}_0, \tag{22}$$

s.t. $\overline{V}_0$ fulfils (20), then we may replace the algebraic equation by an ODE. This explicit ODE for $\overline{V}$ is obtained by differentiation of (7) w.r.t. time, and reads

$$\partial_t \overline{V} = \frac{-\frac{1}{\mathcal{N}_0} \sum_{i=1}^{\mathcal{N}} V_i^{1/3} (\mu_I(\overline{V}, u_1) - \mu_I(V_i, u_1))}{\mathcal{X}(\overline{V}, u_1)(MX_0 - m_1 \frac{1}{\mathcal{N}_0} \sum_{i=1}^{\mathcal{N}(t)} \eta_L(V_i, u_1)\, V_i)} \quad \text{in } [t_0, t_f] \setminus \cup_{1 \leq j \leq \mathcal{N}_0} \{t_j\}, \tag{23}$$

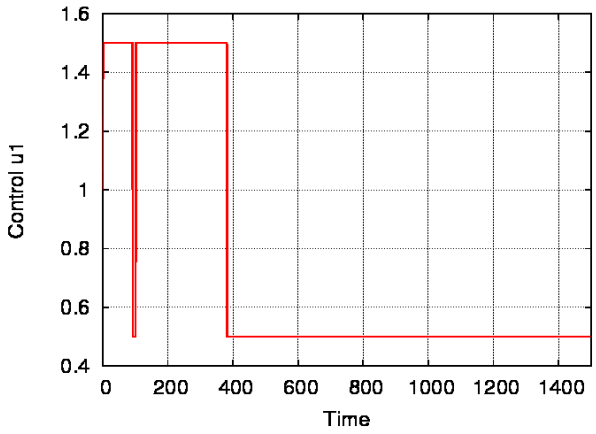

Fig. 2. Control by temperature $[10^2\,\mathrm{K}]$ vs. time $[1\,\mathrm{s}]$

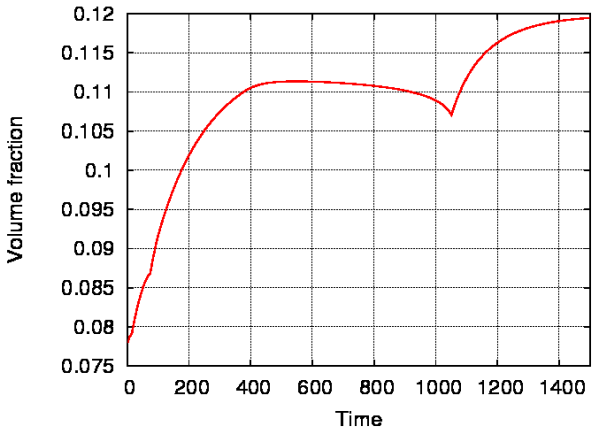

Fig. 3. Volume fraction of precipitates $\frac{1}{\mathcal{N}_0}\sum_{i=1}^{\mathcal{N}_0} V_i(t)$ $[10^{-18}\,\mathrm{m}^3]$ vs. time t $[1\,\mathrm{s}]$

where

$$\mathcal{X}(\overline{V},u_1) = \frac{1}{\eta_S(\overline{V},u_1)} \times \left(\frac{\rho_S(\overline{V},u_1)}{\eta_S(\overline{V},u_1)}\partial_{\overline{V}}\eta_S(\overline{V},u_1) - \partial_{\overline{V}}\rho_S(\overline{V},u_1)\right). \tag{24}$$

After the times t_j when droplets vanish we use (20) in order to determine $\overline{V}(t_j+)$.

We solve our problem by means of

(i) a Runge-Kutta integrator with fixed but suitably small time step size, where the algebraic equation (7) has been replaced by an ODE for $\overline{V}$,
(ii) using the DASSL solver, see Brenan et al. (1996),
  a) keeping the algebraic equation,
  b) replacing the algebraic equation (20) by the ODE $\partial_t V_i = 0$ for $V_i \le V_{min}$ with the initial condition $V_j(t_j+) = 0$.

The algorithms (i), (ii)a) and (ii)b) solve the original problem. However, the algorithms (i) and (ii)b) turn out to run more reliably for large set of initial conditions, while (ii)a) exhibits occasionally problems determining the control at switching points. A critical point with (i) is, that the time step has to be chosen very small for certain data, since the times, when droplets vanish, have to be located as precisely as possible in order to avoid propagated errors. The algorithms based on DASSL turn out to be even more sensitive to the choice of too large time steps. Hence we present our results obtained by algorithm (i) in the following.

3.3 Numerical results

We examine the different contributions to the cost function and the two controls and discuss their impact on the solution. For better illustration, we present our results for radii $r_i = 3/(4\pi)V_i^{1/3}$, corresponding to the special case of spherical precipitates. Fig. 1 shows the time evolution for 5 droplet radii, together with the fictive bulk radius $\overline{r} = 3/(4\pi)\overline{V}^{1/3}$, and Fig. 2 presents the corresponding control. The initial control parameter turns out to be $u_0 = u_{min,0}$. In Fig. 3 we give the evolution of the volume fraction, that enters into the α_2-term of the cost function, but at the final time.

Our numerics suggest a control of bang-bang type for u_1 and that the α_3-term, representing the deviation from the mean droplet volume within the cost functional, has no impact for large times t_f since the mean field $\overline{V}$ represents an unstable stationary point for the V_i. We conjecture that the α_2-term is the most controllable contribution within the cost functional. Furthermore, for sufficiently small time steps our numerical results seem to be mesh independent.

4. THEORETICAL ASPECTS AND OPEN QUESTIONS

Besides further numerical tasks, like more efficient algorithms for long-time behaviour e.g. by a suitable finite-volume discretization for LSW as in Carillo and Goudon (2001), also from a theoretical point of view the above stated optimal control problem exhibits very interesting aspects. The analysis of the classical LSW model without control has been treated in Niethammer and Pego (2000, 2001). Testing the LSW equation (2) yields a non-local hyperbolic conservation law in the dual space $\mathcal{C}$. Within optimal control theory there are several results (e.g. Colombo et al. (2011); Coron et al. (2010); Shang and Wang (2011); Gugat et al. (2006); Jacquet et al. (2006)), for a hyperbolic first-order equation, but as far as known by the author neither non-local conservation laws involving measures as states nor systems with switching between an ODE and an algebraic equation have been considered so far.

We summarize the main distinct features of our problem. We have a measure-valued solution (LSW) or switching conditions (Mean field model), the droplet volume is not bounded from above, but we do not observe shocks as they might occur typically due to nonlinearities of the flux function.

An issue for our optimal control problem is that it depends on the control, how many droplets vanish within a prescribed final time t_f. An adjoint based approach for this hybrid optimal control is an open question where the difficulty is due to the switching conditions. Finally, it would be interesting to consider the control of the inhomogeneous LSW equation where the equation depends weakly on the spatial structure, too.

The theoretical issues of the optimal control of LSW-type models are an important question and are work in progress. Problems of this type have applications also within a wider frame, e.g. highly re-entrant manufacturing systems (Coron et al. (2010)), traffic flow (Benzoni-Gavage et al. (2006)), two phase flow, gas dynamics, or aerospace dynamics.

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