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Abstract

In this work, we derive a new model framework for a porous intercalation electrode with a phase separating active material upon lithium intercalation. We start from a microscopic model consisting of transport equations for lithium ions in an electrolyte phase and intercalated lithium in a solid active phase. Both are coupled through a Neumann–boundary condition modeling the lithium intercalation reaction $Li^+ + e^- \rightleftharpoons Li$. The active material phase is considered to be phase separating upon lithium intercalation. We assume that the porous material is a given periodic microstructure and perform analytical homogenization. Effectively, the microscopic model consists of a diffusion and a Cahn–Hilliard equation, whereas the limit model consists of a diffusion and an Allen–Cahn equation. Thus we observe a Cahn–Hilliard to Allen–Cahn transition during the upscaling process. In the sense of gradient flows, the transition goes in hand with a change in the underlying metric structure of the PDE system.

1 Introduction

The search for new electrode materials is an essential aspect of research and development of lithium ion batteries. In modern intercalation materials, lithium ions are inserted into some solid host material, a mechanism which received the 2019 Nobel price in chemistry [18]. Numerous materials were found or have been developed that allow for some general intercalation reaction $y\,Li^+ + y\,e^- + X \rightleftharpoons Li_y\,X$, for instance $X = CoO_2$ as cathode material or $X = C_6$ as anode material. All intercalation electrodes are essentially an ensemble of intercalation particles, glued or sintered together with some binder material and conductive additives, which yield a porous medium. Due to several shortcomings, for example safety issues, low lithium storage capacity, low resulting cell voltage, market price, availability, environmental aspects or mining conditions of raw materials, the development of next generation battery materials is an ongoing research topic and lead to new materials such as $X= FePO_4$ [23, 14]. Li_yFePO_4 is cheap, environmental friendly, and exhibits a reasonable cell voltage. However, it is a phase separating material, where upon lithium intercalation the material tends to separate into a lithium rich phase, $LiFePO_4$, and a lithium poor phase, $FePO_4$. This property has been subject to a wide scientific discussion [27] and its impact on charging mechanisms, [5], suppression of phase separation [3], many-particle effects [7, 10], and phase separation across particles [21, 28].

In terms of a mathematical model, the phase separating behaviour is encoded in a double-well free energy function $F_{\mathrm{A}}(u_{\mathrm{A}})$, for example the classical Cahn–Hilliard free energy [4]

$$F_{\mathrm{A}}(u_{\mathrm{A}}) = u_{\mathrm{A}} \log u_{\mathrm{A}} + (1-u_{\mathrm{A}}) \log (1-u_{\mathrm{A}}) + a\, u_{\mathrm{A}}(1-u_{\mathrm{A}}) \,, \quad u_{\mathrm{A}} \in (0,1) \tag{1.1}$$

which yields a non-monotone chemical potential function μ_{A} with respect to the lithium content u_{A}, for example (non-dimensionalized with respect to $k_B T$)

$$\mu_{\mathrm{A}}(u_{\mathrm{A}}) = F'_{\mathrm{A}}(u_{\mathrm{A}}) = \log \frac{u_{\mathrm{A}}}{1-u_{\mathrm{A}}} + a(1-2\cdot u_{\mathrm{A}}) \,, \tag{1.2}$$

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which becomes non-monotone for $a > 2$ (see Fig. 1).

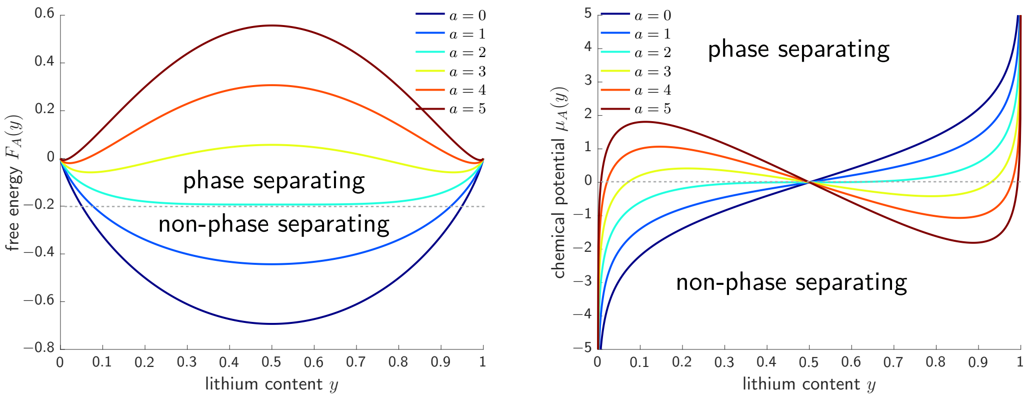

Figure 1: (Left) Free energy density and (right) chemical potential in (1.1) and (1.2) as function of the lithium content y and the phase separation parameter a.

In order to understand, and subsequent control the complex behaviour of a porous, many particle electrode of a phase separating material, precise mathematical models are required. These should be capable of simulating the complex behavior on various degrees of spatial and temporal resolution with predictive value regarding their qualitative and quantitative behaviour. On the other hand, a spatially homogenized model framework for the porous many-particle electrode is desired in order to reduce the numerical degrees of freedom. Such homogenized model frameworks are frequently used for battery modeling, most prominently Newman-type models [17, 6, 11], which, however, neglect phase separation effects.

In this work, we deduce a homogenized model for a periodic porous medium, consisting of an active phase Ω_{A} and an electrolyte phase Ω_{E}. Both are assumed to be simply connected. In the active phase, we consider a rather general Cahn-Hilliard type transport equation for lithium, where $\mu_{\mathrm{A}} = f(u_{\mathrm{A}}) + \gamma_{\mathrm{A}}^{\varepsilon}\mathrm{div}\nabla u_{\mathrm{A}}$, while we assume in the electrolyte a simple diffusion equation for the transport of lithium ions. Of course that later one can be substituted by Poisson-Nernst-Planck type transport equations [8], but for the sake of this work simple diffusion is sufficient. Both phases are connected via a general surface reaction rate, which yields Robin-type conditions at the common interface Σ_{AE}. Due to the porous structure of the domain $\Omega = \Omega_{\mathrm{A}} \cup \Omega_{\mathrm{E}}$, a small scale parameter ε is introduced, which can be considered as the ratio between the diameter of single particle and the porous media width.

Within this setting, we consider a special scaling of the non-equilibrium parameters, namely the diffusion coefficients $D_{\mathrm{A}}^{\varepsilon} = \varepsilon^2 D_{\mathrm{A}}$ and $D_{\mathrm{E}}^{\varepsilon} = \varepsilon^0 D_{\mathrm{E}}$ of the active and the electrolyte phase, respectively, as well as the intercalation reaction rate $R^{\varepsilon} = \varepsilon R$. Most importantly, we consider the interfacial tension term $\gamma_{\mathrm{A}}^{\varepsilon}$ to be in the order of ε^0, i.e. $\gamma_{\mathrm{A}}^{\varepsilon} = \varepsilon^0 \cdot \gamma_{\mathrm{A}}$. For the transient, coupled transport equation system we deduce then the effective, homogenized balance equations, which yield a transition from a Cahn-Hilliard type equation to an Allen-Cahn equation for the active phase. Of course, other scalings of $D_{\mathrm{A}}^{\varepsilon}$ and $\gamma_{\mathrm{A}}^{\varepsilon}$ are thinkable and have partially been studied in literature. We come back to this topic after introducing our model.

2 The root model and related problems from literature

We consider a cuboidal domain $\Omega =]0, \ell_1[\times]0, \ell_2[\times]0, \ell_3[\subset \mathbb{R}^3$ with $\frac{\ell_1}{\ell_2}, \frac{\ell_1}{\ell_3} \in \mathbb{Q}$. For simplicity of presentation, we focus on $\ell_1 = \ell_2 = \ell_3 = 1$.

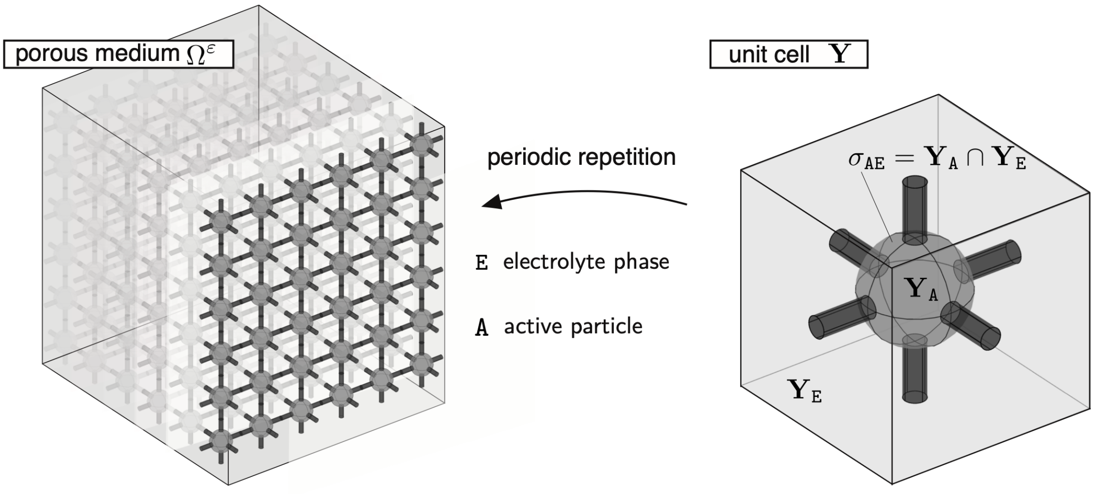

Figure 2: Sketch of the domain.

We decompose the unit cell $\mathbf{Y} := [0,1)^3 = \mathbf{Y}_\mathrm{E} \cup \mathbf{Y}_\mathrm{A}$ into the relatively open sets $\mathbf{Y}_\mathrm{E}$ and $\mathbf{Y}_\mathrm{A}$ with the common interface $\sigma_\mathrm{AE} := \partial\mathbf{Y}_\mathrm{A} \cap \mathbf{Y}_\mathrm{E}$ and we assume that the periodized version of $\mathbf{Y}_\mathrm{E}$ is connected. We then decompose Ω into the (connected) electrolyte domain $\Omega^\varepsilon_\mathrm{E}$ and the (connected) solid domain $\Omega^\varepsilon_\mathrm{A}$ as periodic repetition of the unit cell $\mathbf{Y}_\mathrm{E}$ resp. $\mathbf{Y}_\mathrm{A}$ scaled by ε (see Fig. 2). Moreover, $\Sigma^\varepsilon_\mathrm{AE}$ is the interface between the phase A and E as the periodiced version of $\varepsilon\sigma_\mathrm{AE}$. Without loss of generality, we assume that $\mathrm{Vol}\,\mathbf{Y} = 1$.

On the domain Ω and its components $\Omega^\varepsilon_\mathrm{E}$ and $\Omega^\varepsilon_\mathrm{A}$ we consider the following non-dimensional transport equations:

$$\dot u_\mathrm{A} = \mathrm{div}\big(\varepsilon^2 D_\mathrm{A}\nabla\mu_\mathrm{A}\big) \qquad \text{in } \Omega^\varepsilon_\mathrm{A} \tag{2.1a}$$
$$\mu_\mathrm{A} = -\varepsilon^0\gamma_\mathrm{A}\Delta u_\mathrm{A} + F'_\mathrm{A}(u_\mathrm{A}) \text{in } \Omega^\varepsilon_\mathrm{A} \tag{2.1b}$$
$$\dot u_\mathrm{E} = \mathrm{div}(\varepsilon^0 D_\mathrm{E}\nabla\mu_\mathrm{E}) \qquad \text{in } \Omega^\varepsilon_\mathrm{E} \tag{2.1c}$$
$$\mu_\mathrm{E} = e_\mathrm{E} u_\mathrm{E} \qquad \text{in } \Omega^\varepsilon_\mathrm{A}. \tag{2.1d}$$

Eq. (2.1a) describes the evolution of the concentration field $u_\mathrm{A}(x,t)$ of intercalated lithium in the solid active phase Ω_A. The corresponding chemical potential function μ_A, i.e. eq. (2.1b), is not only dependent on the concentration field u_A, but also on the gradient ∇u_A due to the phase separation effects. In the surrounding electrolyte phase Ω_E we have a (exemplary) the evolution of a single field variable $u_\mathrm{E}(x,t)$, e.g. the concentration of lithium ions, described by (2.1c). For the sake of simplicity we employ a simple chemical potential function μ_E (eq. (2.1d)) as thermodynamic closure relation. The diffusion in the active phase is considered to be small, i.e. in the order of ε^2, while the diffusion in the electrolyte phase is considered to be of order ε^0. Most importantly, however, the phase separation parameter in (2.1b) is considered to be of order ε^0, while classical Cahn-Hilliard homogenization approaches assume orders ε^2 [25].

Both phases are coupled through surface reaction boundary conditions, modeling the intercalation reaction $\mathrm{Li}^+|_\mathrm{E} + \mathrm{e}^-|_\mathrm{A} \rightleftharpoons \mathrm{Li}\,|_\mathrm{A}$. Non-equilibrium surface thermodynamics states [19, 20, 9] a reaction rate $R \propto (\mu_\mathrm{A} - \mu_\mathrm{E})$ such that $R\cdot(\mu_\mathrm{A} - \mu_\mathrm{E}) \geq 0$, which assumes here for the sake of simplicity that the electrochemical potential of the electrons e^- is constant. Most simply we have thus $R = L^\varepsilon \cdot (\mu_\mathrm{A} - \mu_\mathrm{E})$ with L^ε ensuring the local second law of (surface) thermodynamics, and we consider L^ε to be of order ε, i.e. $L^\varepsilon = \varepsilon L$. This yields the following boundary conditions:

$$\varepsilon^2 D_\mathrm{A}\nabla\mu_\mathrm{A}\cdot\nu = -D_\mathrm{E}\nabla\mu_\mathrm{E}\cdot\nu = \varepsilon L\cdot(\mu_\mathrm{A}-\mu_\mathrm{E}) \quad \text{on } \Sigma^\varepsilon_\mathrm{AE} \tag{2.1e}$$

$$\gamma\nabla u_\mathrm{A}\cdot\nu = 0 \quad \text{on } \partial\Omega^\varepsilon_\mathrm{A} \tag{2.1f}$$

$$D_\mathrm{E}\nabla\mu_E\cdot\nu = 0 \quad \text{on } \partial\Omega_\mathrm{E}\backslash\Sigma_\mathrm{AE} \tag{2.1g}$$

$$\varepsilon^2 D_\mathrm{A}\nabla\mu_A\cdot\nu = 0 \quad \text{on } \partial\Omega_\mathrm{A}\backslash\Sigma_\mathrm{AE}\ . \tag{2.1h}$$

Comparison to literature and outlook
We now discuss several obviously related models and generalizations. The first way to generalize our model is to apply different scalings in the parameters. Other generalizations are a disconnectedness of $\Omega^\varepsilon_\mathrm{A}$ and modifications of the constitutive equations of μ_E and the boundary conditions on $\partial\Omega$.

Let us first note that (2.1d) can be replaced by any relation that provides $|\mu'_\mathrm{E}(u_\mathrm{E})| \geq C_{\mu_{\mathrm{E},0}} > 0$. Further, the boundary condition on $\partial\Omega$ is of Robin type in application, which is only a minor modification but makes the work much less readable. Finally, we do not consider disconnected $\Omega^\varepsilon_\mathrm{A}$ because of severe mathematical issues that arise in this case which we were not able to resolve, although we expect that our result would be the same with $A_{\mathrm{hom,A}} = 0$. We come back to this point in the conclusion Section 5.

In case $D^\varepsilon_\mathrm{A} = \varepsilon^0 D_\mathrm{A}$, $\gamma^\varepsilon_\mathrm{A} = \varepsilon^0\gamma_\mathrm{A}$ methods developed in [22] can be applied. The case $D^\varepsilon_\mathrm{A} = \varepsilon^0 D_\mathrm{A}$, $\gamma^\varepsilon_\mathrm{A} = \varepsilon^2\gamma_\mathrm{A}$ was studied in literature [25] with different outcome. Diffusion problems with robin jump conditions on microscopic inclusions have been studied e.g. in [13, 12] and references therein. The work [12] is particularly interesting because it features the effect that the diffusion in phase 2 (in our case phase A) vanishes in the limit and the analysis remains valid also in disconnected phase A. In our case, the fourth order elliptic operator in phase A turns into a second order elliptic operator. However, we note that in case of a disconnected domain $\Omega^\varepsilon_\mathrm{A}$ we expect also this elliptic operator to vanish in the limit.

3 Setting and main result

In this section, we fix the main assumptions for the problem, introduce relevant function spaces as well as necessary constructions, and present the main result. In particular, the crucial assumption is the semiconvexity of the potential F_A.

Assumption 1 (Properties of F_A)*.* We assume that $F_\mathrm{A} : \mathbb{R} \to \mathbb{R}_\infty := \mathbb{R}\cup\{+\infty\}$ is twice differentiable on the interior of its domain and λ-convex with $\lambda \in \mathbb{R}$, which means that $F_0 :\ u \mapsto F_\mathrm{A}(u) - \frac{\lambda}{2}|u|^2$ is convex. Furthermore, *one* of the following two properties are satisfied.

1. There exist constants $-\infty \leq u_- < u_+ \leq +\infty$ such that $\lim_{u\to u_\pm} F_\mathrm{A}(u) = +\infty$.
2. There additionally exists a constant $f_{2,0} \in \mathbb{R}$ such that $\sup_u F''_0(u) < f_{2,0}$.

Remark 2*.* The case $\lambda < 0$ leads to a nonconvex potential F_A. A prototypical example of F_A satisfying Assumption 1.1 is given by the logarithmic double-well potential (see also (1.1))

$$F_\mathrm{A}(u) = \kappa\big((1-u)\log(1-u) + u\log u\big) - \frac{\lambda}{2}u^2. \tag{3.1}$$

We note that every λ-convex functional satisfying Assumption 1.1 can be regularized such that it also satisfies Assumption 1.2. Indeed, assuming $\sup_u F''_\mathrm{A}(u) = +\infty$ consider $u_0 > 0$ and set

$f_{2,0} := 2\sup_{u\in[-u_0,+u_0]} F''_{\mathrm{A}}(u)$, then, the modified function $\tilde F_{\mathrm{A}}$ with $\tilde F''_{\mathrm{A}}(u) = \min\{F''_{\mathrm{A}}(u), f_{2,0}\}$ and $\tilde F_{\mathrm{A}}(0) = F_{\mathrm{A}}(0)$ fulfills the assumption.

3.1 Function spaces

In the following, we write $\mathrm{C}_{\mathrm{per}}(\mathbf{Y})$ for continuous functions on $\mathbb{R}^3$ that are $\mathbf{Y}$-periodic and $\mathrm{C}^k_{\mathrm{per}}(\mathbf{Y})$ for k-times continuous differentiable periodic functions. Furthermore, we write $\mathrm{H}^1_{\mathrm{per}}(\mathbf{Y})$ for $\mathbf{Y}$-periodic functions in $\mathrm{H}^1_{\mathrm{loc}}(\mathbb{R}^3)$. If 'I' stands for either index A or E, we denote by $\mathrm{H}^1_{\mathrm{per}}(\mathbf{Y}_I)$ the restriction of functions in $\mathrm{H}^1_{\mathrm{per}}(\mathbf{Y})$ onto $\mathbf{Y}_{\mathrm{I}}$, and we denote by $\mathrm{H}^1_{\mathrm{per},(0)}(\mathbf{Y}_{\mathrm{I}})$ functions in $\mathrm{H}^1_{\mathrm{per}}(\mathbf{Y}_I)$ having average 0 in $\mathbf{Y}_{\mathrm{I}}$, i.e., $\int_{\mathbf{Y}_{\mathrm{I}}} v(y)\,\mathrm{d}y = 0$

We write $\mathrm{L}^2_{(m)}(A) := \{u \in \mathrm{L}^2(A) \mid \int_A u\,\mathrm{d}x = m\}$ for bounded, open $A \subset \mathbb{R}^d$ and interprete functions on $\Omega^\varepsilon_{\mathrm{I}}$ and $\Omega\times\mathbf{Y}_{\mathrm{I}}$ as functions on Ω resp. on $\Omega\times\mathbf{Y}$ via a simple extension by 0. We define the spaces

$$\mathbf{L}_\varepsilon(\Omega) := \mathrm{L}^2(\Omega^\varepsilon_{\mathrm{A}}) \times \mathrm{L}^2(\Omega^\varepsilon_{\mathrm{E}}) , \quad \mathbf{L}_0(\Omega) := \mathrm{L}^2(\Omega) \times \mathrm{L}^2(\Omega)$$
$$\mathbf{L}(\Omega\times\mathbf{Y}) := \mathrm{L}^2(\Omega\times\mathbf{Y}_{\mathrm{A}}) \times \mathrm{L}^2(\Omega\times\mathbf{Y}_{\mathrm{E}})$$

and identify $v = (v_{\mathrm{A}}, v_{\mathrm{E}}) \in \mathbf{L}_\varepsilon(\Omega)$ with $v = v_{\mathrm{A}} + v_{\mathrm{E}} \in \mathrm{L}^2(\Omega)$ in an isomorphic way and similarly $v = (v_{\mathrm{A}}, v_{\mathrm{E}}) \in \mathbf{L}(\Omega\times\mathbf{Y})$ with $v = \chi_{\mathbf{Y}_{\mathrm{A}}} v_{\mathrm{A}} + \chi_{\mathbf{Y}_{\mathrm{E}}} v_{\mathrm{E}} \in \mathrm{L}^2(\Omega\times\mathbf{Y})$. Then we define

$$\mathbf{L}_{\varepsilon,(m)}(\Omega) := \left\{(u_{\mathrm{A}}, u_{\mathrm{E}}) \in \mathbf{L}_\varepsilon(\Omega) \mid \ \int_\Omega (u_{\mathrm{A}} + u_{\mathrm{E}})\,\mathrm{d}x = m\right\} ,$$
$$\mathbf{L}_{0,(m)}(\Omega) := \left\{(u_{\mathrm{A}}, u_{\mathrm{E}}) \in \mathbf{L}_0(\Omega) \mid \ \int_\Omega (|\mathbf{Y}_{\mathrm{A}}| u_{\mathrm{A}} + |\mathbf{Y}_{\mathrm{E}}| u_{\mathrm{E}})\,\mathrm{d}x = m\right\} .$$

In all cases, we use the notation $v = (v_{\mathrm{A}}, v_{\mathrm{E}})$ to indicate the first and second component. Note that spaces of functions of constant average $m \neq 0$ are only affine spaces. However, when $m = 0$, the above spaces become Hilbert spaces. Furthermore, we introduce the spaces for the chemical potentials

$$\mathbf{H}_\varepsilon(\Omega) := \left(\mathrm{H}^1(\Omega^\varepsilon_{\mathrm{A}}) \times \mathrm{H}^1(\Omega^\varepsilon_{\mathrm{E}})\right) \cap \mathbf{L}_{\varepsilon,(0)}(\Omega) ,$$
$$\mathbf{H}_0(\Omega) := \left(\mathrm{L}^2(\Omega) \times \mathrm{H}^1(\Omega)\right) \cap \mathbf{L}_{0,(0)}(\Omega) .$$

While we use the canonical norms and scalar products on $\mathbf{L}_\varepsilon(\Omega)$ and $\mathbf{L}_0(\Omega)$, we consider the following scalar product on $\mathbf{H}_\varepsilon(\Omega)$

$$\left\langle(\mu_{\mathrm{A}}, \mu_{\mathrm{E}}),\, (\tilde\mu_{\mathrm{A}}, \tilde\mu_{\mathrm{E}})\right\rangle_{\mathbf{H}_\varepsilon(\Omega)} := \int_{\Omega^\varepsilon_{\mathrm{A}}} \varepsilon^2 D_{\mathrm{A}} \nabla\mu_{\mathrm{A}} \cdot \nabla\tilde\mu_{\mathrm{A}}\,\mathrm{d}x + \int_{\Omega^\varepsilon_{\mathrm{E}}} D_{\mathrm{E}} \nabla v_{\mathrm{E}} \cdot \nabla\tilde\mu_{\mathrm{E}}\,\mathrm{d}x$$
$$+ \int_{\Sigma^\varepsilon_{\mathrm{AE}}} \varepsilon L\,(\mu_{\mathrm{A}} - \mu_{\mathrm{E}})\,(\tilde\mu_{\mathrm{A}} - \tilde\mu_{\mathrm{E}})\,\mathrm{d}a\,.$$

The scalar product leads to the canonical norm $\|(\mu_{\mathrm{A}}, \mu_{\mathrm{E}})\|_{\mathbf{H}_\varepsilon(\Omega)}$, which is only a seminorm in the case that $\mathbf{L}_{\varepsilon,(0)}(\Omega)$ is replaced by $\mathbf{L}_{\varepsilon,(m)}(\Omega)$ for $m \neq 0$. For given rates $v = (v_{\mathrm{A}}, v_{\mathrm{E}}) \in \mathbf{L}_\varepsilon(\Omega)$, we define the chemical potentials $\xi^\varepsilon(v) = (\xi^\varepsilon_A(v), \xi^\varepsilon_{\mathrm{E}}(v)) \in \mathbf{H}_\varepsilon(\Omega)$ as the unique minimizer of the following functional on $\mathbf{H}_\varepsilon(\Omega)$:

$$(\xi_{\mathrm{A}}, \xi_{\mathrm{E}}) \mapsto \frac{1}{2}\,\|(\xi_{\mathrm{A}}, \xi_{\mathrm{E}})\|^2_{\mathbf{H}_\varepsilon(\Omega)} - \int_{\Omega^\varepsilon_{\mathrm{A}}} \xi_{\mathrm{A}} v_{\mathrm{A}}\,\mathrm{d}x - \int_{\Omega^\varepsilon_{\mathrm{E}}} \xi_{\mathrm{E}} v_{\mathrm{E}}\,\mathrm{d}x\,. \tag{3.2}$$

Then, for every $\tilde\mu \in \mathbf{H}_\varepsilon(\Omega)$ we find $\langle\xi^\varepsilon(v), \tilde\mu\rangle_{\mathbf{H}_\varepsilon(\Omega)} = \int_{\Omega^\varepsilon_{\mathrm{A}}} \tilde\mu_{\mathrm{A}} v_{\mathrm{A}}\,\mathrm{d}x + \int_{\Omega^\varepsilon_{\mathrm{E}}} \tilde\mu_{\mathrm{E}} v_{\mathrm{E}}\,\mathrm{d}x$, and we have constructed the Riesz isomorphism $\mathcal{J}_\varepsilon : \mathbf{H}_\varepsilon(\Omega)^* \to \mathbf{H}_\varepsilon(\Omega)$ via the closure of $\mathbf{L}_\varepsilon(\Omega)$ with respect to the norm $\|(v_{\mathrm{A}}, v_{\mathrm{E}})\|_{\mathbf{H}_\varepsilon(\Omega)^*} := \|(\xi^\varepsilon_{\mathrm{A}}(v), \xi^\varepsilon_{\mathrm{E}}(v))\|_{\mathbf{H}_\varepsilon(\Omega)}$ such that $\mathcal{J}_\varepsilon v = \xi^\varepsilon(v)$.

Finally, we introduce the spaces

$$\mathbf{V}_{\varepsilon,(m)}(\Omega) := \big(\mathrm{H}^1(\Omega_\mathrm{A}^\varepsilon)\times \mathrm{L}^2(\Omega_\mathrm{E}^\varepsilon)\big)\cap \mathbf{L}_{\varepsilon,(m)}(\Omega)\,,$$
$$\mathbf{V}_{0,(m)}(\Omega) := \big(\mathrm{H}^1(\Omega)\times \mathrm{L}^2(\Omega)\big)\cap \mathbf{L}_{0,(m)}(\Omega)\,.$$

3.2 Cell problems

The derivation of the effective battery model is based on the formulation of cell problems that capture the relevant microscopic behaviour. For the different phases $\mathrm{I}=\mathrm{A},\mathrm{E}$, we introduce the homogenized coefficient matrices $A_{\mathsf{hom},\mathrm{I}}\in\mathbb{R}^{3\times 3}$ via

$$(A_{\mathrm{hom},\mathrm{I}})_{ij} = \frac{1}{|\mathbf{Y}_\mathrm{I}|}\int_{\mathbf{Y}_\mathrm{I}} (e_i - \nabla_y\chi_{\mathrm{I},i})\cdot(e_j-\nabla_y\chi_{\mathrm{I},j})\,\mathrm{d}y,\quad i,j=1,2,3,$$

where $\chi_{\mathrm{I},i}\in \mathrm{H}^1_{\mathrm{per},(0)}(\mathbf{Y}_\mathrm{I})$, for $i=1,2,3$, is the unique solution of the cell problem

$$\forall v\in \mathrm{H}^1_{\mathrm{per},(0)}(\mathbf{Y}_\mathrm{I}):\qquad \int_{\mathbf{Y}_\mathrm{I}}(e_i+\nabla_y\chi_{\mathrm{I},i})\cdot\nabla_y v\,\mathrm{d}y = 0. \tag{3.3}$$

The solution $\widetilde{\xi}_{\mathrm{A},0}\in \mathrm{H}^1_{\mathrm{per}}(\mathbf{Y}_\mathrm{A})$ of the following problem will play an important role:

$$1-\mathrm{div}_y(D_\mathrm{A}\nabla_y\widetilde{\xi}_{\mathrm{A},0}) = 0 \quad\text{on } \mathbf{Y}_\mathrm{A}\,,\qquad -D_\mathrm{A}\nabla_y\widetilde{\xi}_{\mathrm{A},0}\cdot\nu = L\widetilde{\xi}_{\mathrm{A},0}\quad \text{on } \sigma_{\mathrm{AE}}\,. \tag{3.4}$$

Multiplication of the equation by $\widetilde{\xi}_{\mathrm{A},0}$, integration over $\mathbf{Y}_\mathrm{A}$, and integration by parts yields the positive constant

$$c_{\mathrm{A},0} := -\fint_{\mathbf{Y}_\mathrm{A}}\widetilde{\xi}_{\mathrm{A},0}\,\mathrm{d}y = \frac{1}{|\mathbf{Y}_\mathrm{A}|}\bigg(\int_{\mathbf{Y}_\mathrm{A}} D_\mathrm{A}\big|\nabla_y\widetilde{\xi}_{\mathrm{A},0}\big|^2\,\mathrm{d}y + \int_{\sigma_{\mathrm{AE}}} L\widetilde{\xi}_{\mathrm{A},0}^2\,\mathrm{d}a_y\bigg) > 0. \tag{3.5}$$

The scalar product on $\mathbf{H}_0(\Omega)$ is now introduced as

$$\langle(\mu_\mathrm{A},\mu_\mathrm{E}),(\tilde\mu_\mathrm{A},\tilde\mu_\mathrm{E})\rangle_{\mathbf{H}_0(\Omega)} := \int_\Omega D_\mathrm{E}A_{\mathrm{hom},\mathrm{E}}\nabla\mu_\mathrm{E}\cdot\nabla\tilde\mu_\mathrm{E}\,\mathrm{d}x + \int_\Omega \frac{|\mathbf{Y}_\mathrm{A}|}{c_{\mathrm{A},0}}(\mu_\mathrm{A}-\mu_\mathrm{E})(\tilde\mu_\mathrm{A}-\tilde\mu_\mathrm{E})\,\mathrm{d}x\,. \tag{3.6}$$

Again, this scalar product leads to a canonical norm on $\mathbf{H}_0(\Omega)$ given by

$$\|(\mu_\mathrm{A},\mu_\mathrm{E})\|^2_{\mathbf{H}_0} = D_\mathrm{E}\|\nabla\mu_\mathrm{E}\|^2_{A_{\mathsf{hom}}} + \frac{|\mathbf{Y}_\mathrm{A}|}{c_{\mathrm{A},0}}\|\mu_\mathrm{A}-\mu_\mathrm{E}\|^2_{\mathrm{L}^2}.$$

The latter is a seminorm on $\mathbf{H}_{0,(m)}(\Omega)$ for $m\neq 0$. Furthermore, replacing $\mathbf{H}_\varepsilon$ by $\mathbf{H}_0$ in (3.2) we find a Riesz isomorphism $\mathscr{J}_0$ between $\mathbf{H}_0^*(\Omega)$ and $\mathbf{H}_0(\Omega)$.

We note that we have formulated so far three different cell problems, namely (3.3) to obtained the effective coefficient matrices $A_{\mathsf{hom},\mathrm{A}}$ and $A_{\mathsf{hom},\mathrm{E}}$, and (3.4) to introduce $\widetilde{\xi}_{\mathrm{A},0}$ and $c_{\mathrm{A},0}$. All of them will appear in our main homogenization theorem (Theorem 3).

3.3 Main result

We are now in position to state the main result, namely, the convergence of solutions to the system in (2.1) to solutions of an effective limit system. The effective limit system on Ω reads

$$\partial_t u_{\mathrm{A}} = \frac{1}{c_{\mathrm{A},0}} \left(\mu_{\mathrm{E}} - \overline{\mu}_{\mathrm{A}}\right). \tag{3.7a}$$

$$\partial_t u_{\mathrm{E}} = \mathrm{div}(D_{\mathrm{E}} A_{\mathrm{hom,E}} \nabla \mu_{\mathrm{E}}) - \frac{|\mathbf{Y}_{\mathrm{A}}|}{c_{\mathrm{A},0}|\mathbf{Y}_{\mathrm{E}}|} \left(\mu_{\mathrm{E}} - \overline{\mu}_{\mathrm{A}}\right), \tag{3.7b}$$

$$\overline{\mu}_{\mathrm{A}} = F_{\mathrm{A}}'(u_{\mathrm{A}}) - \mathrm{div}\left(\gamma A_{\mathrm{hom,A}} \nabla u_{\mathrm{A}}\right), \tag{3.7c}$$

$$\mu_{\mathrm{E}} = e_{\mathrm{E}} u_{\mathrm{E}} \tag{3.7d}$$

subject to the boundary conditions on $\partial\Omega$

$$\gamma A_{\mathsf{hom},\mathrm{A}} \nabla u_{\mathrm{A}} \cdot \nu = 0\,, \qquad D_{\mathrm{E}} A_{\mathrm{hom,E}} \nabla \mu_{\mathrm{E}} \cdot \nu = 0\,. \tag{3.7e}$$

Theorem 3 (Main Theorem)**.** *Let Ω be as above and assume that either one of the conditions in Assumption 1 is fulfilled.*

(i) For every $\varepsilon > 0$ with $(1/\varepsilon) \in \mathbb{N}$ and initial conditions $u_0^\varepsilon = (u_{0,\mathrm{A}}^\varepsilon, u_{0,\mathrm{E}}^\varepsilon) \in \mathbf{V}_{\varepsilon,(m)}(\Omega)$ satisfying $u_{0,\mathrm{A}}^\varepsilon(x) \in [0,1]$ for a.a. $x \in \Omega_{\mathrm{A}}^\varepsilon$ there exist a unique solution to the problem (2.1) *with $u^\varepsilon = (u_{\mathrm{A}}^\varepsilon, u_{\mathrm{E}}^\varepsilon) \in \mathrm{L}^\infty(0,T;\mathbf{V}_{\varepsilon,(m)}(\Omega))$ with $\mu^\varepsilon = (\mu_{\mathrm{A}}^\varepsilon, \mu_{\mathrm{E}}^\varepsilon) \in \mathrm{L}^2(0,T;\mathbf{H}_\varepsilon(\Omega))$ and $\partial_t u^\varepsilon \in \mathrm{L}^2(0,T;\mathbf{H}_\varepsilon(\Omega)^*)$ and such that $u_{\mathrm{A}}^\varepsilon(x) \in [0,1]$ for a.a. $x \in \Omega_{\mathrm{A}}^\varepsilon$.*

(ii) If the initial data u_0^ε is uniformly bounded in $\mathbf{V}_{\varepsilon,(m)}(\Omega)$, then, the family of solutions u^ε to (2.1) *satisfy the uniform a priori estimates*

$$\|u_{\mathrm{E}}^\varepsilon\|_{\mathrm{L}^\infty(0,T;\mathrm{L}^2(\Omega_{\mathrm{E}}))} + \|u_{\mathrm{A}}^\varepsilon\|_{\mathrm{L}^\infty(0,T;\mathrm{L}^\infty(\Omega_{\mathrm{A}}^\varepsilon))} + \|\nabla u_{\mathrm{A}}^\varepsilon\|_{\mathrm{L}^\infty(0,T;\mathrm{L}^2(\Omega_{\mathrm{A}}^\varepsilon))} \le C\,, \tag{3.8}$$

$$\varepsilon\,\|\nabla \mu_{\mathrm{A}}^\varepsilon\|_{\mathrm{L}^2(0,T;\mathrm{L}^2(\Omega_{\mathrm{A}}^\varepsilon))} + \|\nabla \mu_{\mathrm{E}}^\varepsilon\|_{\mathrm{L}^2(0,T;\mathrm{L}^2(\Omega_{\mathrm{E}}^\varepsilon))} + \sqrt{\varepsilon}\,\|\mu_{\mathrm{A}}^\varepsilon - \mu_{\mathrm{E}}^\varepsilon\|_{\mathrm{L}^2(0,T;\mathrm{L}^2(\Sigma_{\mathrm{AE}}^\varepsilon))} \le C\,, \tag{3.9}$$

$$\left\|\mu_{\mathrm{A}}^{\varepsilon\,2}\right\|_{\mathrm{L}^2(0,T;\mathrm{L}^2(\Omega_{\mathrm{A}}^\varepsilon))} \le C, \tag{3.10}$$

$$\|(\partial_t u_{\mathrm{A}}^\varepsilon, \partial_t u_{\mathrm{E}}^\varepsilon)\|_{\mathrm{L}^2(0,T;\mathbf{H}_\varepsilon^{-1}(\Omega))} \le C, \tag{3.11}$$

where $C > 0$ is a constant independent of ε.

(iii) If $u_0^\varepsilon \xrightarrow{2s} u_0^0$ in two scales (see Definition 5 and (4.7)*), then, the family of solutions u^ε to* (2.1) *also converges in the two-scale sense to the unique solution u to the effective problem in* (3.7)*. The latter satisfies $u \in \mathrm{L}^2(0,T;\mathbf{V}_{0,(m)}(\Omega))$ with $\partial_t u \in \mathrm{L}^2(0,T;\mathbf{H}_0(\Omega)^*)$ and $u(0) = u_0^0$.*

Solutions to (2.1) are gradient flows for the energy functional $\mathcal{E}_\varepsilon : \mathbf{H}_\varepsilon(\Omega)^* \to \mathbb{R}_\infty$ given by

$$\mathcal{E}_\varepsilon(u_{\mathrm{A}}, u_{\mathrm{E}}) = \begin{cases} \displaystyle\int_{\Omega_{\mathrm{A}}^\varepsilon} \frac{\gamma_{\mathrm{A}}}{2}|\nabla u_{\mathrm{A}}|^2 + F_{\mathrm{A}}(u_{\mathrm{A}})\,\mathrm{d}x + \int_{\Omega_{\mathrm{E}}^\varepsilon} \frac{e_{\mathrm{E}}}{2}|u_{\mathrm{E}}|^2\,\mathrm{d}x & \text{if } (u_{\mathrm{A}}, u_{\mathrm{E}}) \in \mathbf{V}_{\varepsilon,(m)}(\Omega), \\ +\infty & \text{otherwise} \end{cases} \tag{3.12}$$

on the Hilbert space $\mathbf{H}_\varepsilon(\Omega)^*$ equipped with the norm $\|\cdot\|_{\mathbf{H}_\varepsilon(\Omega)^*}$, i.e., $\dot{u}^\varepsilon(t) \in \partial_{\mathbf{H}_\varepsilon^*}\mathcal{E}_\varepsilon(u^\varepsilon(t))$, where $\partial_{\mathbf{H}_\varepsilon^*}\mathcal{E}_\varepsilon(u) \subset \mathbf{H}_\varepsilon(\Omega)^*$ denotes the (Fréchet) subdifferential of $\mathcal{E}_\varepsilon$ in u. The functional $\mathcal{E}_\varepsilon$ is lower semicontinuous on $\mathbf{H}_\varepsilon(\Omega)^*$ and has compact sublevels (on $\mathbf{H}_\varepsilon(\Omega)^*$). Moreover, the initial data satisfies $u_\varepsilon^0 \in \mathrm{dom}(\mathcal{E}_\varepsilon)$. Since F_{A} is λ-convex, it can be shown that there exists $\Lambda_\varepsilon \in \mathbb{R}$ such that $\mathcal{E}_\varepsilon$ is Λ_ε-convex (see [22, Lemma 3.7]), in particular, $\mathcal{E}_\varepsilon$ satisfies a chain rule on the Hilbert space $\mathbf{H}_\varepsilon(\Omega)^*$.

Thus, the existence result follows from Theorem 3 in [24] (see also [1]). The uniqueness follows from the Λ_ε-convexity of $\mathcal{E}_\varepsilon$.

The limit system in (3.7) also has a gradient structure. In particular, solutions are gradient flows with respect to the energy functional $\mathcal{E}_0 : \mathbf{H}_0(\Omega)^* \to \mathbb{R}_\infty$ defined as

$$\mathcal{E}_0(u_\mathrm{A}, u_\mathrm{E}) = \begin{cases} \int_\Omega \frac{\gamma_\mathrm{A}}{2}|\nabla u_\mathrm{A}|^2 + F_\mathrm{A}(u_\mathrm{A}) + \frac{e_\mathrm{E}}{2}|u_\mathrm{E}|^2 \,\mathrm{d}x & \text{if } (u_\mathrm{A}, u_\mathrm{E}) \in \mathbf{V}_{0,(m)}(\Omega), \\ +\infty & \text{otherwise.} \end{cases}$$

4 Proof of main result

4.1 Periodic Sobolev theory and two-scale convergence

Lemma 4. *There exists a constant $C > 0$ independent from ε such that*

$$\forall v_\mathrm{E}^\varepsilon \in \mathrm{H}^1(\Omega_\mathrm{E}^\varepsilon) \qquad \int_{\Sigma_\mathrm{AE}^\varepsilon} \varepsilon\,|v_\mathrm{E}^\varepsilon|^2 \,\mathrm{da} \le C \int_{\Omega_\mathrm{E}^\varepsilon} \left(|v_\mathrm{E}^\varepsilon|^2 + \varepsilon^2\,|\nabla v_\mathrm{E}^\varepsilon|^2 \right) \mathrm{d}x, \tag{4.1}$$

$$\forall v_\mathrm{A}^\varepsilon \in \mathrm{H}^1(\Omega_\mathrm{A}^\varepsilon) \qquad \int_{\Omega_\mathrm{A}^\varepsilon} |v_\mathrm{A}^\varepsilon|^2 \,\mathrm{d}x \le C \left(\int_{\Sigma_\mathrm{AE}^\varepsilon} \varepsilon\,|v_\mathrm{A}^\varepsilon|^2 \,\mathrm{da} + \varepsilon^2 \int_{\Omega_\mathrm{A}^\varepsilon} |\nabla v_\mathrm{A}^\varepsilon|^2 \,\mathrm{d}x \right). \tag{4.2}$$

Proof. Note that by assumption $\Omega = (0,1)^3$. For $u \in \mathrm{L}^2(\Omega_\mathrm{A}^\varepsilon)$ (or $u \in \mathrm{L}^2(\Omega_\mathrm{E}^\varepsilon)$) we extend u outside of $\Omega_\mathrm{A}^\varepsilon$ (or $\Omega_\mathrm{E}^\varepsilon$) and consider iteratively:

1. for $x = (x_1, x_2, x_3) \in (0,1)^3$ the function $u(-x_1, x_2, x_3) := u(x_1, x_2, x_3)$,
2. for $x = (x_1, x_2, x_3) \in (-1,1) \times (0,1)^2$ the function $u(x_1, -x_2, x_3) := u(x_1, x_2, x_3)$,
3. for $x = (x_1, x_2, x_3) \in (-1,1)^2 \times (0,1)$ the function $u(x_1, x_2, -x_3) := u(x_1, x_2, x_3)$.

In particular, u can be continued to a $(-1,1)^3$-periodic function with the property that $u \in \mathrm{H}^1(\Omega_\mathrm{A}^\varepsilon)$ implies H^1-regularity for the extended function and similarly for $u \in \mathrm{H}^1(\Omega_\mathrm{E}^\varepsilon)$. Using this extension, the estimates follow from periodicity and a scaling argument. □

Next, we establish the uniform boundedness of the solutions to (2.1), which follows from the energy-dissipation balance.

Proof of a priori bounds. Note that solutions u^ε to (2.1) are gradient flows in $\mathbf{H}_\varepsilon(\Omega)^*$ with respect to $\mathcal{E}_\varepsilon$ according to Theorem 3 in [24]. In particular, they satisfy the energy-dissipation balance

$$\mathcal{E}_\varepsilon(u^\varepsilon(t)) + \int_0^t \int_{\Omega_\mathrm{A}^\varepsilon} \varepsilon^2 D_\mathrm{A} |\nabla \mu_\mathrm{A}^\varepsilon|^2 \,\mathrm{d}x\,\mathrm{d}s + \int_0^t \int_{\Omega_\mathrm{E}^\varepsilon} D_\mathrm{E} |\nabla \mu_\mathrm{E}^\varepsilon|^2 \,\mathrm{d}x\,\mathrm{d}s + \int_0^t \int_{\Sigma_\mathrm{AE}} \varepsilon L (\mu_\mathrm{A}^\varepsilon - \mu_\mathrm{E}^\varepsilon)^2 \,\mathrm{da}\,\mathrm{d}s = \mathcal{E}_\varepsilon(u_{0,\mathrm{A}}^\varepsilon, u_{0,\mathrm{E}}^\varepsilon). \tag{4.3}$$

Since, we assume well-preparedness of the initial condition, i.e., $\mathcal{E}_\varepsilon(u_{0,\mathrm{A}}^\varepsilon, u_{0,\mathrm{E}}^\varepsilon) \le C < \infty$ the estimates in (3.8) and (3.9) follow directly. The scaled trace inequalities (4.1) and (4.2) then imply (3.10). The estimate for the time derivatives in (3.11) follows from (3.9) via standard arguments.

□

Since $\Omega_{\mathrm{E}}^{\varepsilon}$ and $\Omega_{\mathrm{A}}^{\varepsilon}$ are connected, it is known (see [16, 15] and references therein) that there exists $C_{\mathcal{U},\mathrm{I}} > 0$ such that for every $\varepsilon = 1/n$, $n \in \mathbb{N}$, there exists an extension operator $\mathcal{U}_{\mathrm{I}}^{\varepsilon} : \mathrm{H}^1(\Omega_{\mathrm{I}}^{\varepsilon}) \to \mathrm{H}^1(\mathbb{R}^3)$ such that

$$\begin{aligned} \forall u \in \mathrm{H}^1(\Omega_{\mathrm{I}}^{\varepsilon}): \quad & (\mathcal{U}_{\mathrm{I}}^{\varepsilon} u)|_{\Omega_{\mathrm{I}}^{\varepsilon}} = u, \quad \|\mathcal{U}_{\mathrm{I}}^{\varepsilon} u\|_{\mathrm{L}^2(\Omega)} \leq C_{\mathcal{U},\mathrm{I}} \|u\|_{\mathrm{L}^2(\Omega_{\mathrm{I}}^{\varepsilon})}, \\ \text{and} \quad & \|\nabla(\mathcal{U}_{\mathrm{I}}^{\varepsilon} u)\|_{\mathrm{L}^2(\Omega)}^2 \leq C_{\mathcal{U},\mathrm{I}} \|\nabla u\|_{\mathrm{L}^2(\Omega_{\mathrm{I}}^{\varepsilon})}^2 . \end{aligned} \tag{4.4}$$

Next, we observe that the trace operator $\mathcal{T} : \mathrm{H}^1(\mathbf{Y}_{\mathrm{I}}) \to \mathrm{L}^2(\sigma_{\mathrm{AE}})$ is well defined and continuous for both $\mathrm{I} \in \{\mathrm{A}, \mathrm{E}\}$ with continuity constant $C_{\mathcal{T},\mathrm{I}}$. From a simple scaling argument this implies that

$$\mathcal{T}^{\varepsilon} : \begin{cases} \mathrm{H}^1(\Omega_{\mathrm{I}}^{\varepsilon}) & \to \quad \mathrm{L}^2(\Sigma_{\mathrm{AE}}^{\varepsilon}), \\ \quad u & \mapsto \quad [\mathcal{T} u(\varepsilon\, \cdot)] \left(\frac{\cdot}{\varepsilon}\right) \end{cases}$$

is continuous with the estimate

$$\|\mathcal{T}^{\varepsilon} u\|_{\mathrm{L}^2(\Sigma_{\mathrm{AE}}^{\varepsilon})} \leq C_{\mathcal{T},\mathrm{I}} \left(\|u\|_{\mathrm{L}^2(\Omega_{\mathrm{I}}^{\varepsilon})} + \varepsilon \|\nabla u\|_{\mathrm{L}^2(\Omega_{\mathrm{I}}^{\varepsilon})}\right) . \tag{4.5}$$

Definition 5 (Two-scale convergence, [29])**.** (i) We call a (Radon) measure ω on $\mathbb{R}^3$ periodic if for every $z \in \mathbb{Z}^3$ and every open subset $A \subset \mathbb{R}^3$ it holds $\omega(A) = \omega(A + z)$. Given a periodic measure ω_0 we define for $\varepsilon > 0$ the family of scaled measures $\omega_{\varepsilon}(A) := \varepsilon^3 \omega_0(\varepsilon^{-1} A)$.

(ii) Given a periodic measure ω_0 on $\mathbb{R}^3$ and the associated family of scaled measures ω_{ε} for every $\varepsilon > 0$, we say that a sequence $u_{\varepsilon} \in \mathrm{L}^2(\Omega; \omega_{\varepsilon})$ converges weakly in two scales to $u \in \mathrm{L}^2(\Omega; \mathrm{L}^2(\mathbf{Y}; \omega_0))$ if $\sup_{\varepsilon>0} \|u_{\varepsilon}\|_{\mathrm{L}^2(\Omega;\omega_{\varepsilon})} < \infty$ and for every $\phi \in \mathrm{C}(\overline{\Omega})$ and $\psi \in \mathrm{L}^2(\mathbf{Y})$ it holds

$$\lim_{\varepsilon \to 0} \int_{\Omega} u_{\varepsilon}(x)\, \phi(x) \psi\left(\frac{x}{\varepsilon}\right) \mathrm{d}\omega_{\varepsilon}(x) = \int_{\Omega} \int_{\mathbf{Y}} u(x, y)\, \phi(x) \psi(y)\, \mathrm{d}\omega_0(y)\, \mathrm{d}x \,.$$

Remark 6. While in [2] the space $\mathrm{L}^2(\Omega; \mathrm{C}_{\mathrm{per}}(\mathbf{Y}))$ is preferred as set of test functions, it is noticed therein that also other type of test functions would be possible, which are classified as 'admissible'. This includes the space $\mathrm{L}^2(\mathbf{Y}; \mathrm{C}(\overline{\Omega}))$. However, in [29] it is revealed that the only important ingredient into the theory is the density of the test functions in $\mathrm{L}^2(\Omega; \mathrm{L}^2(\mathbf{Y}; \omega_0))$.

The most important result which makes the theory applicable is the following.

Proposition 7 ([29, Prop. 2.2])**.** *If a sequence u_{ε} is bounded in $\mathrm{L}^2(\Omega; \omega_{\varepsilon})$, then it is precompact in the sense of two-scale convergence.*

Remark 8. In this work, we consider the two cases for the periodic measure ω: (i) $\omega_0 = \mathcal{L}^3$, the three-dimensional Lebesgue measure, such that $\omega_{\varepsilon} = \mathcal{L}^3$ and (ii) $\omega_{0,\sigma}(A) := \mathcal{H}^2(\sigma_{\mathrm{AE}} \cap A)$, the two-dimensional Hausdorff measure such that $\omega_{\varepsilon,\Sigma}(A) = \omega_{\sigma}^{\varepsilon}(A) := \varepsilon \mathcal{H}^2(\Sigma_{\mathrm{AE}}^{\varepsilon} \cap A)$. Since the characteristic functions χ_{I} of $\mathbf{Y}_{\mathrm{I}}$, for $\mathrm{I} = \mathrm{A}, \mathrm{E}$, are periodic, also $\omega_{0,\mathrm{I}} = \chi_{\mathrm{I}} \omega_0$ is a periodic measure with associated scaled measures $\omega_{\varepsilon,\mathrm{I}} = \chi_{\mathrm{I}}(\frac{\cdot}{\varepsilon})\mathcal{L}^3$. It is straight forward that $u_{\varepsilon} \xrightarrow{2s} u$ with respect to ω_{ε} implies $u_{\varepsilon}\chi_{\mathrm{I}}(\frac{\cdot}{\varepsilon}) \xrightarrow{2s} \chi_{\mathrm{I}} u$ with respect to $\omega_{\varepsilon,\mathrm{I}}$.

In the case $\omega_0 = \mathcal{L}^3$, we find the following important results.

Lemma 9 ([2, Prop. 1.14])**.** *Let u_{ε} be a sequence in $\mathrm{H}^1(\Omega)$ such that for $\alpha \in \{0, 1\}$*

$$\sup_{\varepsilon>0} \left\{ \|u_{\varepsilon}\|_{\mathrm{L}^2(\Omega)} + \varepsilon^{\alpha} \|\nabla u_{\varepsilon}\|_{\mathrm{L}^2(\Omega)} \right\} < \infty \,.$$

Then, there exists $u \in \mathrm{L}^2(\Omega \times \mathbf{Y})$ such that along a subsequence $u_{\varepsilon} \xrightarrow{2s} u$. Furthermore depending on α the following holds:

(i) If $\alpha = 0$, then $u \in \mathrm{H}^1(\Omega)$, i.e., $u(x,y) = u(x)$ does not depend on y and there exists $v \in \mathrm{L}^2(\Omega; \mathrm{H}^1_{\mathrm{per}}(\mathbf{Y}))$ such that $\nabla u_\varepsilon \xrightarrow{2s} \nabla u + \nabla_y v$.

(ii) If $\alpha = 1$, then $u \in \mathrm{L}^2(\Omega; \mathrm{H}^1_{\mathrm{per}}(\mathbf{Y}))$ and $\varepsilon \nabla u_\varepsilon \xrightarrow{2s} \nabla_y u$.

The last lemma can be used to prove the following result, which has been obtained in similar formulations before (see e.g. [13] and references therein). We provide a proof for the sake of self-containedness.

Lemma 10. *Let $u_\varepsilon : \Omega^\varepsilon_{\mathrm{I}} \to \mathbb{R}$ be a sequence in $\mathrm{H}^1(\Omega^\varepsilon_{\mathrm{I}})$, for $\mathrm{I} \in \{\mathrm{A}, \mathrm{E}\}$, such that*

$$\sup_{\varepsilon > 0} \left\{ \|u_\varepsilon\|_{\mathrm{L}^2(\Omega^\varepsilon_{\mathrm{I}})} + \varepsilon \|\nabla u_\varepsilon\|_{\mathrm{L}^2(\Omega^\varepsilon_{\mathrm{I}})} \right\} < \infty.$$

Then, there exists $u \in \mathrm{L}^2(\Omega; \mathrm{H}^1_{\mathrm{per}}(\mathbf{Y}))$ such that along a subsequence

(i) $u_\varepsilon \xrightarrow{2s} \chi_{\mathrm{I}} u$ and $\varepsilon \nabla u_\varepsilon \xrightarrow{2s} \chi_{\mathrm{I}} \nabla_y u$ with respect to the scaled measures $\omega_{\varepsilon,\mathrm{I}} = \chi_{\mathrm{I}}(\frac{\cdot}{\varepsilon})\mathcal{L}^3$,

(ii) $\mathcal{U}^\varepsilon_{\mathrm{I}} u_\varepsilon \xrightarrow{2s} u$ and $\varepsilon \nabla (\mathcal{U}^\varepsilon_{\mathrm{I}} u_\varepsilon) \xrightarrow{2s} \nabla_y u$ with respect to the scaled measure $\omega = \mathcal{L}^3$,

(iii) $\mathcal{T}^\varepsilon u_\varepsilon \xrightarrow{2s} \mathcal{T} u$ with respect to the scaled measure $\omega^\varepsilon_\sigma = \varepsilon \mathcal{H}^2(\Sigma^\varepsilon_{\mathrm{AE}} \cap A)$.

Proof. We define $\tilde{u}^\varepsilon := \mathcal{U}^\varepsilon_{\mathrm{I}} u_\varepsilon$. Due to (4.4) as well as Lemma 9, there exists $u \in \mathrm{L}^2(\Omega; \mathrm{H}^1_{\mathrm{per}}(\mathbf{Y}))$ such that along a subsequence $\tilde{u}^\varepsilon \xrightarrow{2s} u$, $\varepsilon \nabla \tilde{u}^\varepsilon \xrightarrow{2s} \nabla_y u$. By Remark 8 and the identity $u_\varepsilon = \chi_{\mathrm{I}}(\frac{\cdot}{\varepsilon}) \mathcal{U}^\varepsilon_{\mathrm{I}} u_\varepsilon$, it follows that $u_\varepsilon \xrightarrow{2s} \chi_{\mathrm{I}} u$ and $\varepsilon \nabla u_\varepsilon \xrightarrow{2s} \chi_I \nabla_y u$. Estimate (4.5) yields that $\mathcal{T}^\varepsilon u_\varepsilon$ is two-scale precompact, and it remains to identify the limit.

We write ν^ε for the outer normal vector of $\Omega^\varepsilon_{\mathrm{I}}$ on $\Sigma^\varepsilon_{\mathrm{AE}}$ and ν for the outer normal vector of $\mathbf{Y}_{\mathrm{I}}$ on σ_{AE}. Then, the set $\{\psi \cdot \nu : \psi \in \mathrm{C}^1(\mathbf{Y}_{\mathrm{I}})^d\}$ is dense in $\mathrm{L}^2(\sigma_{\mathrm{AE}})$, and it is sufficient to use $\psi \cdot \nu$ as test functions for two-scale convergence, where $\psi \in \mathrm{C}^1(\mathbf{Y}_{\mathrm{I}})$. Indeed, we then observe that

$$\begin{aligned} \int_{\Sigma^\varepsilon_{\mathrm{AE}}} \mathcal{T}^\varepsilon u_\varepsilon(x)\, \nu^\varepsilon(x) \cdot \psi(\tfrac{x}{\varepsilon})\, \mathrm{d}\omega^\varepsilon_\sigma(x) &= \int_{\Omega^\varepsilon_{\mathrm{I}}} \varepsilon \mathrm{div} \left(u_\varepsilon(x) \psi(\tfrac{x}{\varepsilon}) \right) \mathrm{d}x \\ &\to \int_\Omega \int_{\mathbf{Y}_{\mathrm{I}}} \mathrm{div}_y \left(u(x,y) \psi(y) \right) \mathrm{d}x\, \mathrm{d}y = \int_{\sigma_{\mathrm{AE}}} \mathcal{T} u(x,y)\, \psi(y) \cdot \nu \, \mathrm{d}a\,, \end{aligned}$$

which proves that $\mathcal{T}^\varepsilon u_\varepsilon \xrightarrow{2s} \mathcal{T} u$ with respect to the scaled measure $\omega^\varepsilon_\sigma$. □

4.2 Limit passage

Lemma 11. *The sequences $\mathcal{U}^\varepsilon_{\mathrm{E}} u^\varepsilon_{\mathrm{E}}$ and $\mathcal{U}^\varepsilon_{\mathrm{A}} u^\varepsilon_{\mathrm{A}}$ are precompact in $\mathrm{L}^2([0,T] \times \Omega)$.*

Proof. We focus on the sequence $\mathcal{U}^\varepsilon_{\mathrm{E}} u^\varepsilon_{\mathrm{E}}$ the case for $\mathcal{U}^\varepsilon_{\mathrm{A}} u^\varepsilon_{\mathrm{A}}$ follows analogously. We verify the applicability of Simon's theorem [26, Thm. 1]. For that, we need to show that (i) for all $0 \le t_1 < t_2 \le T$ the function $U^\varepsilon_{\mathrm{E}}(x) := \int_{t_1}^{t_2} \mathcal{U}^\varepsilon_{\mathrm{E}} u^\varepsilon_{\mathrm{E}}(s,x)\, \mathrm{d}s$ is precompact in $\mathrm{L}^2(\Omega)$ and that (ii) $\mathcal{U}^\varepsilon_{\mathrm{E}} u^\varepsilon_{\mathrm{E}}(\cdot + h) \to \mathcal{U}^\varepsilon_{\mathrm{E}} u^\varepsilon_{\mathrm{E}}$ in $\mathrm{L}^2([0, T{-}h] \times \Omega)$ as $h \to 0$ uniformly with respect to ε. Concerning the first point, let us write $\tilde{u}^\varepsilon_{\mathrm{E}} := \mathcal{U}^\varepsilon_{\mathrm{E}} u^\varepsilon_{\mathrm{E}}$. The properties of the extension operator in (4.4) yield

$$\|\tilde{u}^\varepsilon_{\mathrm{E}}\|^2_{\mathrm{L}^2([0,T]\times\Omega)} + \|\nabla \tilde{u}^\varepsilon_{\mathrm{E}}\|^2_{\mathrm{L}^2([0,T]\times\Omega)} \le C\big(\|u^\varepsilon_{\mathrm{E}}\|^2_{\mathrm{L}^2([0,T]\times\Omega^\varepsilon_{\mathrm{E}})} + \|\nabla u^\varepsilon_{\mathrm{E}}\|^2_{\mathrm{L}^2([0,T]\times\Omega^\varepsilon_{\mathrm{E}})} \big). \tag{4.6}$$

Since we have $\sup_{\varepsilon>0} \|U_{\rm E}^\varepsilon\|_{{\rm H}^1(\Omega)} \leq C$ due to the a priori estimates for $u_{\rm E}^\varepsilon$ in (3.8) and for $\mu_{\rm E}^\varepsilon = e_0 u_{\rm E}^\varepsilon$ in (3.9), the precompactness of $U_{\rm E}^\varepsilon$ follows from the compact embedding of ${\rm H}^1(\Omega)$ into ${\rm L}^2(\Omega)$. For the second condition, we set $w_{{\rm E},h}^\varepsilon(t) := u_{\rm E}^\varepsilon(t{+}h) - u_{\rm E}^\varepsilon(t)$ and obtain

$$\begin{aligned}\int_0^{T-h} \big\|w_{{\rm E},h}^\varepsilon\big\|^2_{{\rm L}^2(\Omega_{\rm E}^\varepsilon)} \,{\rm d}t &\leq \int_0^{T-h}\int_t^{t+h} \big\langle \partial_t u_{\rm E}^\varepsilon(s), w_{{\rm E},h}^\varepsilon(t)\big\rangle \,{\rm d}s\,{\rm d}t \\ &= \int_0^{T-h}\int_t^{t+h}\int_{\Sigma_{\rm AE}^\varepsilon} \varepsilon L\big(\mu_{\rm E}^\varepsilon(s){-}\mu_{\rm A}^\varepsilon(s)\big) w_{{\rm E},h}^\varepsilon(t)\,{\rm d}a\,{\rm d}s\,{\rm d}t \\ &\quad + \int_0^{T-h}\int_t^{t+h}\int_{\Omega_{\rm E}^\varepsilon} D_{\rm E}\nabla\mu_{\rm E}^\varepsilon(s)\cdot\nabla w_{\rm E}^\varepsilon(t)\,{\rm d}x\,{\rm d}s\,{\rm d}t,\end{aligned}$$

where we used the evolution equation (2.1c) for $u_{\rm E}^\varepsilon$.

Now, we observe that for $t \in (0, T{-}h)$ Hölder's inequality for $g \in {\rm L}^2(0,T)$

$$\left(\int_t^{t+h} g(s)\,{\rm d}s\right)^2 \leq h\int_t^{t+h} g(s)^2\,{\rm d}s \leq h\|g\|^2_{{\rm L}^2(0,T)}$$

Hence, for $g(s) = \|\mu_{\rm E}^\varepsilon(s){-}\mu_{\rm A}^\varepsilon(s)\|_{{\rm L}^2(\Sigma_{\rm AE}^\varepsilon)}$ and $g(s) = \|\nabla\mu_{\rm E}^\varepsilon(s)\|_{{\rm L}^2(\Omega_{\rm E}^\varepsilon)}$ using the a priori estimates (3.8) and (3.9) and the inequality (4.1), we get with property (4.4) of the extension operator $\mathcal{U}_{\rm E}^\varepsilon$

$$\int_0^{T-h} \|\tilde u_{\rm E}^\varepsilon(\cdot+h) - \tilde u_{\rm E}^\varepsilon(\cdot)\|^2_{{\rm L}^2(\Omega)}\,{\rm d}t \leq C\int_0^{T-h}\|w_{\rm E}^\varepsilon(\cdot)\|^2_{{\rm L}^2(\Omega_{\rm E}^\varepsilon)}\,{\rm d}t \leq \sqrt{h}C\,.$$

This finishes the proof and the precompactness of $\tilde u_{\rm E}^\varepsilon := \mathcal{U}_{\rm E}^\varepsilon u_{\rm E}^\varepsilon$ in ${\rm L}^2([0,T]{\times}\Omega)$ is established. □

We are now in position to probe the two-scale convergence of the solutions. Note that the a priori estimates in (3.8)–(3.11) and points (i) and (ii) of Lemma 10 allow us to find subsequences and limits $u_{\rm E} \in {\rm L}^2(0,T;{\rm H}^1(\Omega))$ and $v_{\rm E} \in {\rm L}^2(0,T;{\rm L}^2(\Omega;{\rm H}^1_{\rm per}(\mathbf{Y})))$ satisfying

$$\mathcal{U}_{\rm E}^\varepsilon u_{\rm E}^\varepsilon \rightharpoonup u_{\rm E} \quad \text{in } {\rm L}^2(0,T;{\rm H}^1(\Omega)), \qquad \nabla(\mathcal{U}_{\rm E}^\varepsilon u_{\rm E}^\varepsilon) \xrightharpoonup{2s} \nabla u_{\rm E} + \nabla_y v_{\rm E} \tag{4.7a}$$

$$u_{\rm E}^\varepsilon \xrightharpoonup{2s} \chi_{\rm E} u_{\rm E}, \qquad \nabla u_{\rm E}^\varepsilon \xrightharpoonup{2s} \chi_{\rm E}\left(\nabla u_{\rm E} + \nabla_y v_{\rm E}\right), \tag{4.7b}$$

where the convergences in (4.7b) are with respect to the scaled measure $\omega_{{\rm E},\varepsilon} = \chi_{\rm E}(\cdot/\varepsilon)\mathcal{L}^3$. Due to Lemma 11, we can further assume that $\mathcal{U}_{\rm E}^\varepsilon u_{\rm E}^\varepsilon \to u_{\rm E}$ strongly in ${\rm L}^2([0,T]{\times}\Omega))$ and Lemma 10 gives the convergence of the traces with respect to the scaled measure $\omega_\sigma^\varepsilon = \varepsilon\mathcal{H}^2(\Sigma_{\rm AE}^\varepsilon \cap A)$:

$$\mathcal{T}^\varepsilon \mu_{\rm E}^\varepsilon \xrightharpoonup{2s} \mathcal{T}\mu_{\rm E}\,. \tag{4.7c}$$

Furthermore, there exists $\mu_{\rm A} \in {\rm L}^2(0,T;{\rm L}^2(\Omega;{\rm H}^1_{\rm per}(\mathbf{Y}_{\rm A})))$ such that

$$\mu_{\rm A}^\varepsilon \xrightharpoonup{2s} \mu_{\rm A}\,, \qquad \varepsilon\nabla\mu_{\rm A}^\varepsilon \xrightharpoonup{2s} \nabla_y\mu_{\rm A}\,. \tag{4.7d}$$

Concerning the convergence of $u_{\rm A}^\varepsilon$ let us note that since $\Omega_{\rm A}^\varepsilon$ is connected there exist a limit $u_{\rm A} \in {\rm L}^2(0,T;{\rm H}^1(\Omega))$, $v_{\rm A} \in {\rm L}^2(0,T;{\rm L}^2(\Omega;{\rm H}^1_{\rm per}(\mathbf{Y})))$ such that

$$\mathcal{U}_{\rm A}^\varepsilon u_{\rm A}^\varepsilon \rightharpoonup u_{\rm A} \quad \text{in } {\rm L}^2(0,T;{\rm H}^1(\Omega))\,, \qquad \nabla(\mathcal{U}_{\rm A}^\varepsilon u_{\rm A}^\varepsilon) \xrightharpoonup{2s} \nabla u_{\rm A} + \nabla_y v_{\rm A}, \tag{4.7e}$$

$$u_{\rm A}^\varepsilon \xrightharpoonup{2s} \chi_{\rm A} u_{\rm A}\,, \qquad \nabla u_{\rm A}^\varepsilon \xrightharpoonup{2s} \chi_{\rm A}\left(\nabla u_{\rm A} + \nabla_y v_{\rm A}\right)\,. \tag{4.7f}$$

Furthermore, by Lemma 11 we get that $\mathcal{U}_{\rm A}^\varepsilon u_{\rm A}^\varepsilon \to u_{\rm A}$ strongly in $\mathrm{L}^2(0,T;\mathrm{L}^2(\Omega))$ and Lemma 10 gives the convergence of the traces with respect to the scaled measure $\omega_\sigma^\varepsilon = \varepsilon\mathcal{H}^2(\Sigma_{\rm AE}^\varepsilon \cap A)$:

$$\mathcal{T}^\varepsilon \mu_{\rm A}^\varepsilon \stackrel{2s}{\rightharpoonup} \mathcal{T}\mu_{\rm A}\,. \tag{4.7g}$$

For the time derivatives $\partial_t u_{\rm E}^\varepsilon$ and $\partial_t u_{\rm A}^\varepsilon$, we consider test functions $v_{\rm E} \in \mathrm{C}_0^1(0,T;\mathrm{H}^1(\Omega))$ and $v_{\rm A} \in \mathrm{C}_0^1(0,T;\mathrm{C}^1(\overline{\Omega}\times\mathbf{Y}_{\rm A}))$ being periodic in $\mathbf{Y}$. We set $\tilde v_{\rm A}^\varepsilon(t,x) := v_{\rm A}(t,x,x/\varepsilon)$. From the a priori estimate (3.11), we infer

$$\begin{aligned}\int_0^T \left|\left\langle(\partial_t u_{\rm A}^\varepsilon,\partial_t u_{\rm E}^\varepsilon),(\tilde v_{\rm A}^\varepsilon, v_{\rm E})\right\rangle\right|\,\mathrm{d}t \le C\Big(&\left\|\nabla_x v_{\rm E}\right\|_{\mathrm{L}^2([0,T]\times\Omega)} + \varepsilon\left\|\nabla_x v_{\rm A}\right\|_{\mathrm{L}^2([0,T]\times\Omega\times\mathbf{Y})}\\ &+\left\|\nabla_y v_{\rm A}\right\|_{\mathrm{L}^2([0,T]\times\Omega\times\mathbf{Y})} + \sqrt{\varepsilon}\left\|v_{\rm A}-v_{\rm E}\right\|_{\mathrm{L}^2([0,T]\times\Sigma_{\rm AE}^\varepsilon)}\Big)\,.\end{aligned}$$

In the limit $\varepsilon\to 0$ we recall (3.6) to obtain the inequality

$$\begin{aligned}\limsup_{\varepsilon\to0}\int_0^T \left|\left\langle(\partial_t u_{\rm A}^\varepsilon,\partial_t u_{\rm E}^\varepsilon),(\tilde v_{\rm A}^\varepsilon, v_{\rm E})\right\rangle\right|\,\mathrm{d}t \le C\Big(&\left\|\nabla_x v_{\rm E}\right\|_{\mathrm{L}^2([0,T]\times\Omega)}\\ &+\left\|\nabla_y v_{\rm A}\right\|_{\mathrm{L}^2([0,T]\times\Omega\times\mathbf{Y})} + \left\|v_{\rm A}-v_{\rm E}\right\|_{\mathrm{L}^2([0,T]\times\Omega\times\Sigma_{\rm AE})}\Big)\end{aligned} \tag{4.8}$$

On the other hand, we can obtain from the fact that both limits $u_{\rm E}$ and $u_{\rm A}$ are independent from y via integration by parts that

$$\begin{aligned}\int_0^T \left\langle(\partial_t u_{\rm A}^\varepsilon,\partial_t u_{\rm E}^\varepsilon),(\tilde v_{\rm A}^\varepsilon, v_{\rm E})\right\rangle\mathrm{d}t &= -\int_0^T\int_{\Omega_{\rm E}^\varepsilon} u_{\rm E}^\varepsilon\,\partial_t v_{\rm E}\,\mathrm{d}x\,\mathrm{d}t + \int_0^T\int_{\Omega_{\rm A}^\varepsilon} u_{\rm A}^\varepsilon\,\partial_t\tilde v_{\rm A}^\varepsilon\,\mathrm{d}x\,\mathrm{d}t\\ &\to -\int_0^T\int_\Omega u_{\rm E}\,\partial_t v_{\rm E}\,\mathrm{d}x\,\mathrm{d}t - \int_0^T\int_\Omega\int_{\mathbf{Y}_{\rm A}} u_{\rm A}(t,x)\partial_t v_{\rm A}(t,x,y)\,\mathrm{d}y\,\mathrm{d}x\,\mathrm{d}t.\end{aligned}$$

We now stick to $v_{\rm E}\in\mathrm{C}_0^1(0,T;\mathrm{H}^1(\Omega))$ but restrict to test functions $v_{\rm A}\in\mathrm{C}_0^1(0,T;\mathrm{C}^1(\overline{\Omega}))$ and obtain from (4.8)

$$\limsup_{\varepsilon\to0}\left|\int_0^T\left\langle(\partial_t u_{\rm E}^\varepsilon,\partial_t u_{\rm A}^\varepsilon),(v_{\rm E},v_{\rm A})\right\rangle\mathrm{d}t\right| \le C\left\|v\right\|_{\mathrm{L}^2(0,T;\mathbf{H}_0(\Omega))}\,.$$

This implies that the time derivatives $(\chi_{\Omega_{\rm E}^\varepsilon}\partial_t u_{\rm E}^\varepsilon, \chi_{\Omega_{\rm A}^\varepsilon}\partial_t u_{\rm A}^\varepsilon)$ converge weakly as distributions to a limit in the space $\mathrm{L}^2(0,T;\mathbf{H}_0(\Omega))$. More precisely, we infer that

$$\partial_t(\chi_{\Omega_{\rm E}^\varepsilon}u_{\rm E}^\varepsilon,\chi_{\Omega_{\rm A}^\varepsilon}u_{\rm A}^\varepsilon) \rightharpoonup \partial_t(|\mathbf{Y}_{\rm E}|u_{\rm E},|\mathbf{Y}_{\rm A}|u_{\rm A})\quad\text{weakly in } \mathrm{L}^2(0,T;\mathbf{H}_0(\Omega))\,. \tag{4.9}$$

Note that the properties of the subdifferential $\partial_{\mathbf{H}_\varepsilon^*}\mathcal{E}_\varepsilon(u^\varepsilon)$ yields that we have that $F_{\rm A}'(u_{\rm A}^\varepsilon)$ is uniformly bounded in $\mathrm{L}^2([0,T]\times\Omega_{\rm A}^\varepsilon)$. Thus, we can also assume that $\mathcal{U}_{\rm A}^\varepsilon F_{\rm A}'(u_{\rm A}^\varepsilon)\rightharpoonup\xi_{\rm A}$ in $\mathrm{L}^2([0,T]\times\Omega)$. Since $u_{\rm A}\mapsto F_{\rm A}(u_{\rm A})$ is λ-convex, we get with the strong convergence of $\mathcal{U}_{\rm A}^\varepsilon u_{\rm A}^\varepsilon$ from Lemma 11 and the strong-weak closedness of the convex subdifferential that for all $\phi\in\mathrm{L}^2([0,T]\times\Omega)$

$$\int_0^T\int_{\Omega_{\rm A}^\varepsilon}\big(F_{\rm A}'(u_{\rm A}^\varepsilon)-\lambda u_{\rm A}^\varepsilon\big)\phi\,\mathrm{d}x\,\mathrm{d}t \to \int_0^T\int_\Omega\big(F_{\rm A}'(u_{\rm A})-\lambda u_{\rm A}\big)\phi\,\mathrm{d}x\,\mathrm{d}t. \tag{4.10}$$

We finally consider the convergence of the initial data. For this aim we chose test functions $v_{\mathrm{E}} \in \mathrm{C}^1([0,T];\mathrm{H}^1(\Omega))$ and $v_{\mathrm{A}} \in \mathrm{C}^1([0,T];\mathrm{C}^1(\overline{\Omega}))$ with $(v_{\mathrm{A}}(T), v_{\mathrm{E}}(T)) = (0,0)$ and use integration by parts to find the following limit behavior:

$$\begin{aligned}\int_\Omega \big\{|\mathbf{Y}_{\mathrm{A}}|u^0_{0,\mathrm{A}}v_{\mathrm{A}}(0) + |\mathbf{Y}_{\mathrm{E}}|u^0_{0,\mathrm{E}}v_{\mathrm{E}}(0)\big\}\,\mathrm{d}x &= \lim_{\varepsilon\to 0}\left(\int_{\Omega^\varepsilon_{\mathrm{A}}} u^\varepsilon_{0,\mathrm{A}}v_{\mathrm{A}}(0)\,\mathrm{d}x + \int_{\Omega^\varepsilon_{\mathrm{E}}} u^\varepsilon_{0,\mathrm{E}}v_{\mathrm{E}}(0)\,\mathrm{d}x\right)\\
&= \lim_{\varepsilon\to 0}\int_0^T\left(\int_{\Omega^\varepsilon_{\mathrm{A}}}\partial_t(u^\varepsilon_{\mathrm{A}}(t)v_{\mathrm{A}}(t))\,\mathrm{d}x + \int_{\Omega^\varepsilon_{\mathrm{E}}}\partial_t(u^\varepsilon_{\mathrm{E}}(t)v_{\mathrm{E}}(t))\,\mathrm{d}x\right)\mathrm{d}t\\
&= \int_0^T\left(\int_\Omega |\mathbf{Y}_{\mathrm{A}}|\partial_t(u_{\mathrm{A}}(t)v_{\mathrm{A}}(t))\,\mathrm{d}x + \int_\Omega |\mathbf{Y}_{\mathrm{E}}|\partial_t(u_{\mathrm{E}}(t)v_{\mathrm{E}}(t))\,\mathrm{d}x\right)\mathrm{d}t\\
&= \int_\Omega\big\{|\mathbf{Y}_{\mathrm{A}}|u_{\mathrm{A}}(0)v_{\mathrm{A}}(0) + |\mathbf{Y}_{\mathrm{E}}|u_{\mathrm{E}}(0)v_{\mathrm{E}}(0)\big\}\,\mathrm{d}x.\end{aligned}$$

Homogenization limit in equations. We are now in position to pass to the limit using the "classical" approach in the equations for $u^\varepsilon_{\mathrm{A}}$ and $u^\varepsilon_{\mathrm{E}}$. For simplicity of notation, we identify $\mathcal{T}\mu_{\mathrm{A}}$ with μ_{A} and $\mathcal{T}\mu_{\mathrm{E}}$ with μ_{E}.

Testing (2.1b) with $\varphi_\varepsilon \in \mathrm{C}^1(\overline{\Omega})$ given via $\varphi_\varepsilon(x) := \phi_0(x) + \varepsilon\phi_1(x)\psi(\frac{x}{\varepsilon})$, where $\phi_0,\phi_1 \in \mathrm{C}^1(\overline{\Omega})$, $\psi \in \mathrm{H}^1(\mathbf{Y}_{\mathrm{A}})$, using the boundary condition in (2.1f) gives the weak formulation

$$\begin{aligned}\int_0^T\int_{\Omega^\varepsilon_{\mathrm{A}}}\mu^\varepsilon_{\mathrm{A}}\varphi_\varepsilon\,\mathrm{d}x\,\mathrm{d}t &= \int_0^T\int_{\Omega^\varepsilon_{\mathrm{A}}} F'_{\mathrm{A}}(u^\varepsilon_{\mathrm{A}})\varphi_\varepsilon\,\mathrm{d}x\,\mathrm{d}t\\
&\quad + \int_0^T\int_{\Omega^\varepsilon_{\mathrm{A}}}\gamma\nabla u^\varepsilon_{\mathrm{A}}(t,x)\cdot\nabla\left(\phi_0(x) + \varepsilon\phi_1(x)\psi\Big(\frac{x}{\varepsilon}\Big)\right)\mathrm{d}x\,\mathrm{d}t\,.\end{aligned}$$

Exploiting the convergence in (4.7d) and (4.7f) as well as (4.10) gives in the two-scale limit

$$\begin{aligned}\int_0^T\int_\Omega\int_{\mathbf{Y}_{\mathrm{A}}}\phi_0(x)\mu_{\mathrm{A}}(t,x,y)\,\mathrm{d}y\,\mathrm{d}x\,\mathrm{d}t &= |\mathbf{Y}_{\mathrm{A}}|\int_0^T\int_\Omega F'_{\mathrm{A}}(u_{\mathrm{A}}(t,x))\phi_0(x)\,\mathrm{d}x\\
&\quad + \int_0^T\int_\Omega\int_{\mathbf{Y}_{\mathrm{A}}}\gamma\big(\nabla u_{\mathrm{A}}(t,x) + \nabla_y v_{\mathrm{A}}(t,x,y)\big)\cdot\big(\nabla\phi_0(x) + \phi_1(x)\nabla_y\psi(y)\big)\,\mathrm{d}y\,\mathrm{d}x\,\mathrm{d}t\,.\end{aligned}$$

It is well known that for given u_{A} the function $v_{\mathrm{A}} := \sum_{i=1}^3 \partial_i u_{\mathrm{A}}\chi_{\mathrm{A},i}$, where $\chi_{\mathrm{A},i}$ are given via the cell problem (3.3), is the only valid choice. Hence, the last equation takes the canonical form

$$\int_0^T\int_\Omega\int_{\mathbf{Y}_{\mathrm{A}}}\phi_0\mu_{\mathrm{A}}\,\mathrm{d}y\,\mathrm{d}x\,\mathrm{d}t = |\mathbf{Y}_{\mathrm{A}}|\int_0^T\int_\Omega\Big\{F'_{\mathrm{A}}(u_{\mathrm{A}})\phi_0 + \nabla u_{\mathrm{A}}\cdot\gamma A_{\mathrm{hom,A}}\nabla\phi_0\Big\}\,\mathrm{d}x\,\mathrm{d}t.$$

The latter is formally equivalent to the strong formulation

$$\overline{\mu}_{\mathrm{A}} := \frac{1}{|\mathbf{Y}_{\mathrm{A}}|}\int_{\mathbf{Y}_{\mathrm{A}}}\mu_{\mathrm{A}}\,\mathrm{d}y = F'_{\mathrm{A}}(u_{\mathrm{A}}) - \mathrm{div}\left(\gamma A_{\mathrm{hom,A}}\nabla u_{\mathrm{A}}\right)\,. \tag{4.11}$$

Let us now consider the equation for u_{A} in (2.1a). We test the latter with $\psi_\varepsilon(t,x) := \phi_1(t,x)\psi(\frac{x}{\varepsilon})$, where $\phi_1 \in \mathrm{C}^1([0,T]\times\overline{\Omega})$, $\psi \in \mathrm{H}^1(\mathbf{Y}_{\mathrm{A}})$ with $\phi(0,\cdot) = \phi(T,\cdot) = 0$ and use the boundary condition (2.1e) to obtain

$$\int_0^T\int_{\Omega^\varepsilon_{\mathrm{A}}}\Big\{-u^\varepsilon_{\mathrm{A}}\partial_t\psi_\varepsilon + \varepsilon^2 D_{\mathrm{A}}\nabla\mu^\varepsilon_{\mathrm{A}}\cdot\nabla\psi_\varepsilon\Big\}\,\mathrm{d}x\,\mathrm{d}t + \int_0^T\int_{\Sigma^\varepsilon_{\mathrm{AE}}}\varepsilon L(\mu^\varepsilon_{\mathrm{A}} - \mu^\varepsilon_{\mathrm{E}})\psi_\varepsilon\,\mathrm{d}a\,\mathrm{d}t = 0.$$

With the convergences in (4.7c), (4.7d), (4.7f), and (4.7g) this identity yields in the limit $\varepsilon \to 0$

$$\int_0^T \int_\Omega \int_{\mathbf{Y}_\mathrm{A}} \Big\{ -u_\mathrm{A}\psi\partial_t\phi_1 + \phi_1 \nabla_y \mu_\mathrm{A} D_\mathrm{A} \nabla_y \psi \Big\} \,\mathrm{d}y\,\mathrm{d}x\,\mathrm{d}t \\ + \int_0^T \int_\Omega \int_{\sigma_\mathrm{AE}} L(\mu_\mathrm{A} - \mu_\mathrm{E})\phi_1\psi \,\mathrm{d}a_y\,\mathrm{d}x\,\mathrm{d}t = 0,$$

Thus, for almost every $x \in \Omega$ we have that u_A and μ_A are weak solutions to

$$\begin{aligned} \partial_t u_\mathrm{A} - \mathrm{div}_y(D_\mathrm{A}\nabla_y\mu_\mathrm{A}) &= 0 && \text{on } \mathbf{Y}_\mathrm{A}, \\ -D_\mathrm{A}\nabla_y\mu_\mathrm{A}\cdot\nu &= L(\mu_\mathrm{A} - \mu_\mathrm{E}) && \text{on } \sigma_\mathrm{AE}\,. \end{aligned} \tag{4.12}$$

Next, we pass to the limit in equation (2.1c). Using a similar procedure as in the previous step with test function $\varphi_\varepsilon(t,x) := \phi_0(t,x) + \varepsilon\phi_1(t,x)\psi(\frac{x}{\varepsilon})$, where $\phi_0, \phi_1 \in \mathrm{C}^1([0,T]\times\overline{\Omega})$, $\psi \in \mathrm{H}^1(\mathbf{Y}_\mathrm{E})$ leads to

$$\int_0^T \int_\Omega \int_{\mathbf{Y}_\mathrm{E}} \partial_t u_\mathrm{E}\phi_0 \,\mathrm{d}y\,\mathrm{d}x\,\mathrm{d}t + |\mathbf{Y}_\mathrm{E}| \int_0^T \int_\Omega \nabla\mu_\mathrm{E}\cdot A_{\mathrm{hom,E}} D_\mathrm{E}\nabla\phi_0 \,\mathrm{d}x\,\mathrm{d}t \\ = -\int_0^T \int_\Omega \int_{\sigma_\mathrm{AE}} L(\mu_\mathrm{E} - \mu_\mathrm{A})\phi_0 \,\mathrm{d}a_y\,\mathrm{d}x\,\mathrm{d}t,$$

where we have used the convergences in (4.7b), (4.7c), and (4.7g).

Thus, we obtain that u_E and $\mu_\mathrm{E} = e_\mathrm{E} u_\mathrm{E}$ satisfy

$$\partial_t u_\mathrm{E} - \mathrm{div}_x(A_{\text{hom,E}} D_\mathrm{E}\nabla\mu_\mathrm{E}) = \frac{L}{|\mathbf{Y}_\mathrm{E}|}\int_{\sigma_\mathrm{AE}} (\mu_A(\cdot,\cdot,y) - \mu_\mathrm{E})\,\mathrm{d}a_y$$

Dimension reduction. Using $\widetilde{\xi}_0$ from the cell problem (3.4) and the fact that (4.9) implies $\partial_t u_\mathrm{A} \in \mathrm{L}^2([0,T]\times\Omega)$ as well as the fact that $\mu_\mathrm{E} \in \mathrm{L}^2([0,T]\times\Omega)$, we observe that $\widehat{\mu}_\mathrm{A} := \partial_t u_\mathrm{A}\widetilde{\xi}_0 + \mu_\mathrm{E}$ is an element of $\mathrm{L}^2(0,T;\mathrm{L}^2(\Omega;\mathrm{H}^1_\mathrm{per}(\mathbf{Y})))$ and solves (4.12). Indeed, since μ_E does not depend on y, we compute

$$-\mathrm{div}_y(D_\mathrm{A}\nabla_y\widehat{\mu}_\mathrm{A}) = -\partial_t u_\mathrm{A}\mathrm{div}_y(D_\mathrm{A}\nabla_y\widetilde{\xi}_0) = -\partial_t u_\mathrm{A}.$$

For the boundary flux, we have

$$-D_\mathrm{A}\nabla_y\widehat{\mu}_\mathrm{A}\cdot\nu = -D_\mathrm{A}\partial_t u_\mathrm{A}\nabla_y\widetilde{\xi}_0\cdot\nu = L\partial_t u_\mathrm{A}\widetilde{\xi}_0 = L(\widehat{\mu}_\mathrm{A} - \mu_\mathrm{E}).$$

Moreover, with the definition of $c_{\mathrm{A},0}$ in (3.5), we find that $\widehat{\mu}_\mathrm{A} = \partial_t u_\mathrm{A}\widetilde{\xi}_0 + \mu_\mathrm{E}$ also satisfies

$$\frac{1}{|\mathbf{Y}_\mathrm{A}|}\int_{\mathbf{Y}_\mathrm{A}} \widehat{\mu}_\mathrm{A}\,\mathrm{d}y = -c_{\mathrm{A},0}\partial_t u_\mathrm{A} + \mu_\mathrm{E}$$

Inserting (4.11) now yields $\partial_t u_\mathrm{A} = \frac{1}{c_{\mathrm{A},0}}(\mu_\mathrm{E} - \overline{\mu}_\mathrm{A})$ or

$$\partial_t u_\mathrm{A} = \frac{1}{c_{\mathrm{A},0}}\big[\mu_\mathrm{E} - F'_\mathrm{A}(u_\mathrm{A}) + \mathrm{div}\,(\gamma A_{\mathrm{hom,E}}\nabla u_\mathrm{A})\big]. \tag{4.13}$$

Thus, we have proven the effective limit equation for u_A.

We use Gauss' theorem for (4.12) to get

$$\int_{\sigma_{\mathrm{AE}}} L(\mu_{\mathrm{E}} - \mu_{\mathrm{A}})\,\mathrm{da}_y = \int_{\mathbf{Y}_{\mathrm{A}}} \partial_t u_{\mathrm{A}}\,\mathrm{d}y = \frac{|\mathbf{Y}_{\mathrm{A}}|}{c_{\mathrm{A},0}}\,(\mu_{\mathrm{E}} - \overline{\mu}_{\mathrm{A}})$$

and obtain

$$|\mathbf{Y}_{\mathrm{E}}|\,\partial_t u_{\mathrm{E}} = |\mathbf{Y}_{\mathrm{E}}|\,\mathrm{div}(D_{\mathrm{E}} A_{\mathrm{hom,E}} \nabla \mu_{\mathrm{E}}) - \frac{|\mathbf{Y}_{\mathrm{A}}|}{c_{\mathrm{A},0}}\,(\mu_{\mathrm{E}} - \overline{\mu}_{\mathrm{A}})\ . \tag{4.14}$$

This finally proves Theorem 3, i.e., the convergence of the solutions u^ε to solutions of the effective problem.

5 Conclusion

As already stated in the introduction, an open issue remains the case of disconnected $\Omega_{\mathrm{A}}^\varepsilon$, which is relevant for application. Conceptually, there does not seem to be any deep reason, why a result similar to the homogenization result in Theorem 3 should not hold in this case with $A_{\mathrm{hom,A}} = 0$. In particular, recent results in [12] underpin this stand point. However, from the point of view of analytical homogenization, the major issue is the lack of compactness of $\mathcal{U}_{\mathrm{A}}^\varepsilon u_{\mathrm{A}}^\varepsilon$ in $\mathrm{L}^2(0,T;\mathrm{L}^2(\Omega))$. Compared to the recent work [12], this lack is due to the fact that the Cahn–Hilliard equation forces us to compare solutions with their shifted versions in H^{-1} instead of L^2. Worse, the space $\mathrm{H}^{-1}(\Omega_{\mathrm{A}}^\varepsilon)$ depends in our case on ε, as we have seen, and the limit problem is again an Allen–Cahn type problem in the space L^2. We were not able to identify a way to compensate for this issue.

However, in order to formally justify our educated guess of the limit equations in the disconnected case, note that disconnectedness of $\mathbf{Y}_{\mathrm{A}}$ implies $A_{\mathrm{hom}} = 0$ (since in this case for every i the function $\chi_{\mathrm{A},i}(y) = -y_i$ is the unique solution of the cell problem). We can test (3.7c) by u_{A} and find

$$\int_\Omega \overline{\mu}_{\mathrm{A}} u_{\mathrm{A}}\,\mathrm{d}x \geq \int_\Omega F_{\mathrm{A}}'(u_{\mathrm{A}}) u_{\mathrm{A}}\,\mathrm{d}x + \int_\Omega \gamma A_{\mathrm{hom,A}} |\nabla u_{\mathrm{A}}|^2\,\mathrm{d}x$$

and from there

$$\frac{1}{2}\int_\Omega \overline{\mu}_{\mathrm{A}}^2\,\mathrm{d}x + C \geq \frac{1}{2}\int_\Omega |u_{\mathrm{A}}|^2\,\mathrm{d}x + \int_\Omega \gamma A_{\mathrm{hom,A}} |\nabla u_{\mathrm{A}}|^2\,\mathrm{d}x\,.$$

This implies for the weak formulation

$$\int_\Omega \overline{\mu}_{\mathrm{A}} \phi\,\mathrm{d}x = \int_\Omega F_{\mathrm{A}}'(u_{\mathrm{A}})\phi\,\mathrm{d}x + \sqrt{A_{\mathrm{hom,A}}}\int_\Omega \gamma\left(\sqrt{A_{\mathrm{hom,A}}}\nabla u_{\mathrm{A}}\right)\cdot\nabla\phi\,\mathrm{d}x$$

in the limit $A_{\mathrm{hom,A}} \to 0$ that

$$\int_\Omega \overline{\mu}_{\mathrm{A}} \phi\,\mathrm{d}x = \int_\Omega F_{\mathrm{A}}'(u_{\mathrm{A}})\phi\,\mathrm{d}x\,.$$

This is a weak formulation of

$$\overline{\mu}_{\mathrm{A}}(x)\,\mathrm{d}y = F_{\mathrm{A}}'(u_{\mathrm{A}})\,.$$

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