

# Next-generation all-solid-state battery

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## Mathematical modelling for the next-generation All-solid-state batteries: Nucleation (SE|SSE)<sup>(\*)</sup>-interface

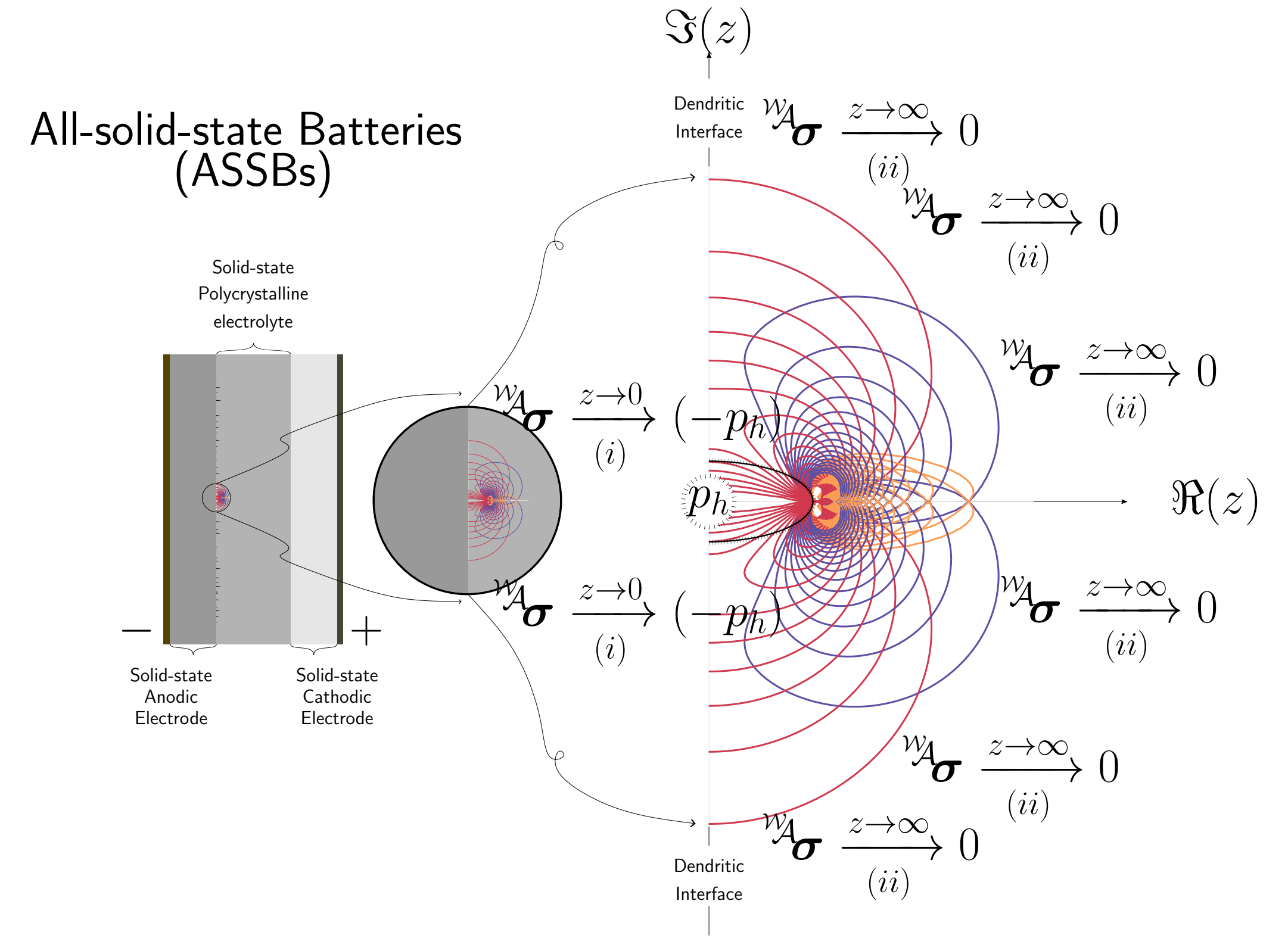
**Rechargeable Lithium-ion battery (LIB)** is at the heart of every electric vehicle (EV), portable electronic device, and energy storage system [1]. Nowadays, LIBs enable human life more efficient and help to solve global environment issues thanks to EVs' zero emission. However, conventional LIB (c-LIB) is sensible to temperature and pressure, hence, flammable and explosive. This bottleneck is mainly due to liquid-based electrolyte in c-LIBs.

**All-solid-state battery (ASSB)** is one of promising candidates to overcome bottlenecks of c-LIBs. Thanks to solid-state electrolyte (SSE), ASSB is highly stable towards temperature and pressure. Nevertheless, metallic Li-dendrite triggered at (SE|SSE)-interface is the main drawback as these dendritic threads extrapolate into grain boundary network of SSE, causing crevice, degradation of ionic conductivity, and the probability of short-circuit.

**Next-generation All-solid-state battery (ng-ASSB)** with a consideration of nucleation criterion defined by

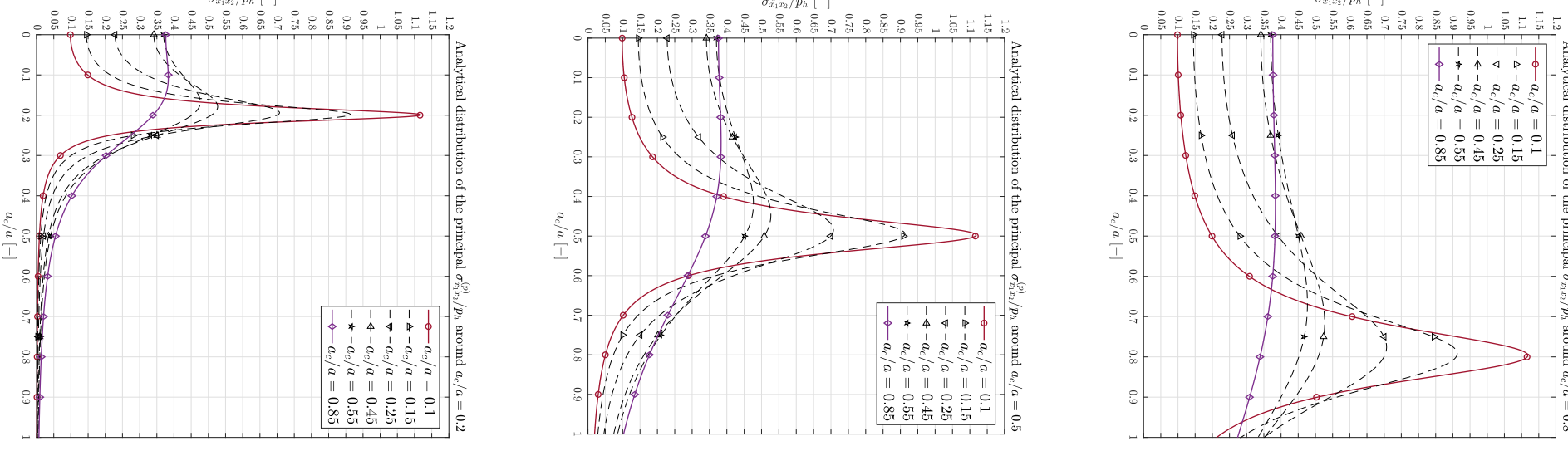
$$a_{\text{Griffith}} := a^* = \arg \min_{a \in \mathbb{R}} \iint_{\Omega} f(a, \mathbf{u}; \lambda, \mu, \mathbf{d} \otimes \mathbf{d}) d\Omega - \iint_{\Gamma} f(a; \gamma) d\Gamma \Big|_{\mathbf{u}^{(s)}}$$

where, can help to improve ASSB performance.



### Interface

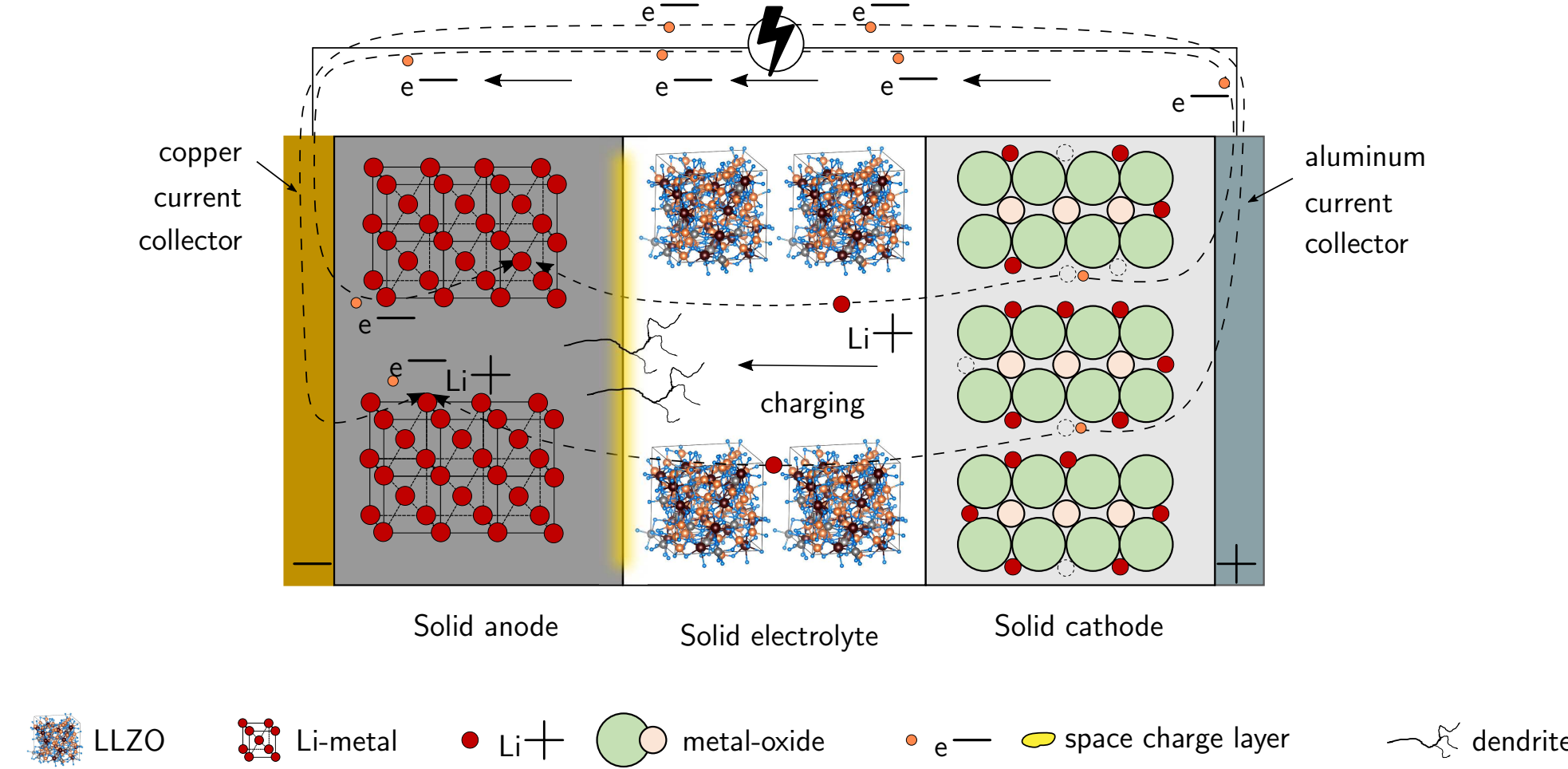
**Interface** between solid electrode and solid-state electrolyte (SE|SSE) taking place at space charge layer (SCL) [2] found in ASSBs critically exhibits mechanical and electrochemical instability [3]. This evidence points directly to the fact that the soft metallic Li anode is erroneously prone to triggering dendrites, under cycles of electric charge & discharge [4].



Distribution: ana. max. shear stress  $\frac{w_1}{\sigma} \Pi_{x_1 x_2}$  around crack tip  $a_c$ .

### Next-generation All-solid-state battery

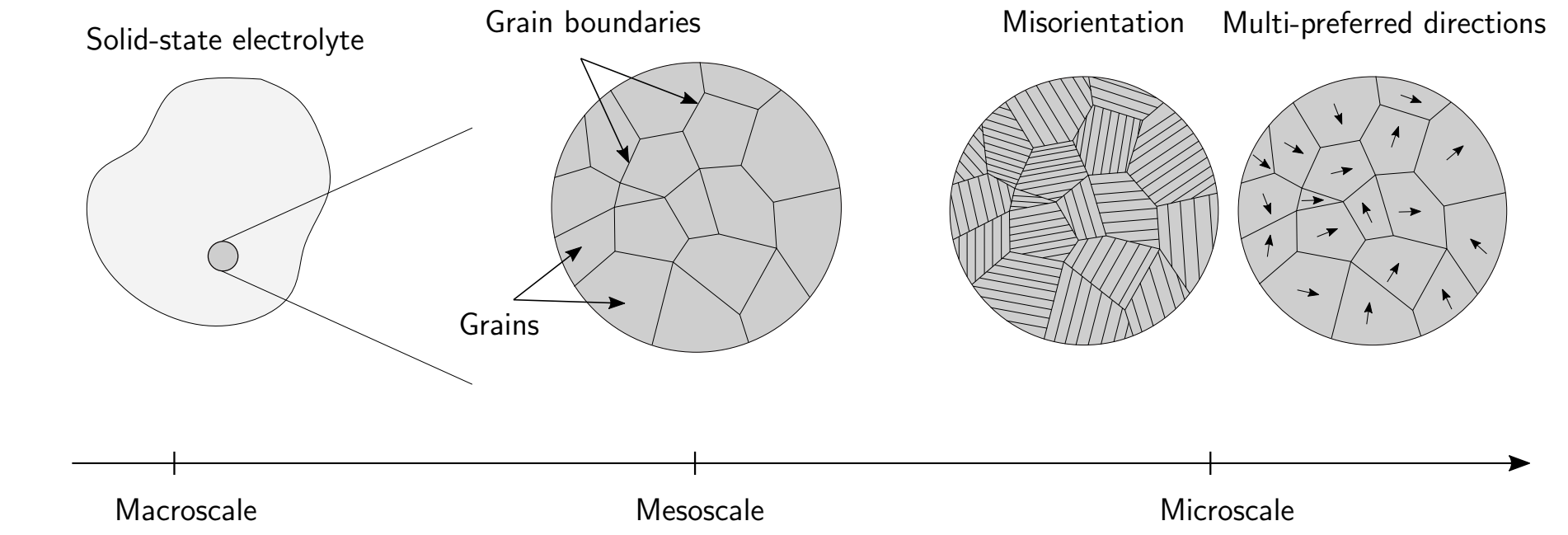
Nucleation taking place at critical dendritic (SE|SSE)-interface



Nucleation taking place at critical dendritic (SE|SSE)-interface  
Nucleation taking place at critical dendritic (SE|SSE)-interface

### Embedded structural-tensor SSE

Polycrystalline garnet-typed SSE such as LLZO exhibit grain boundaries and various sizes and shapes of grains under microscopic observation. Therefore, this type of microstructure distinctively leads to nuance destruction of ceramic-like materials.



Consequently, dendrites contribute to degradation of ionic conductivity and trace along grain boundaries in SSE.

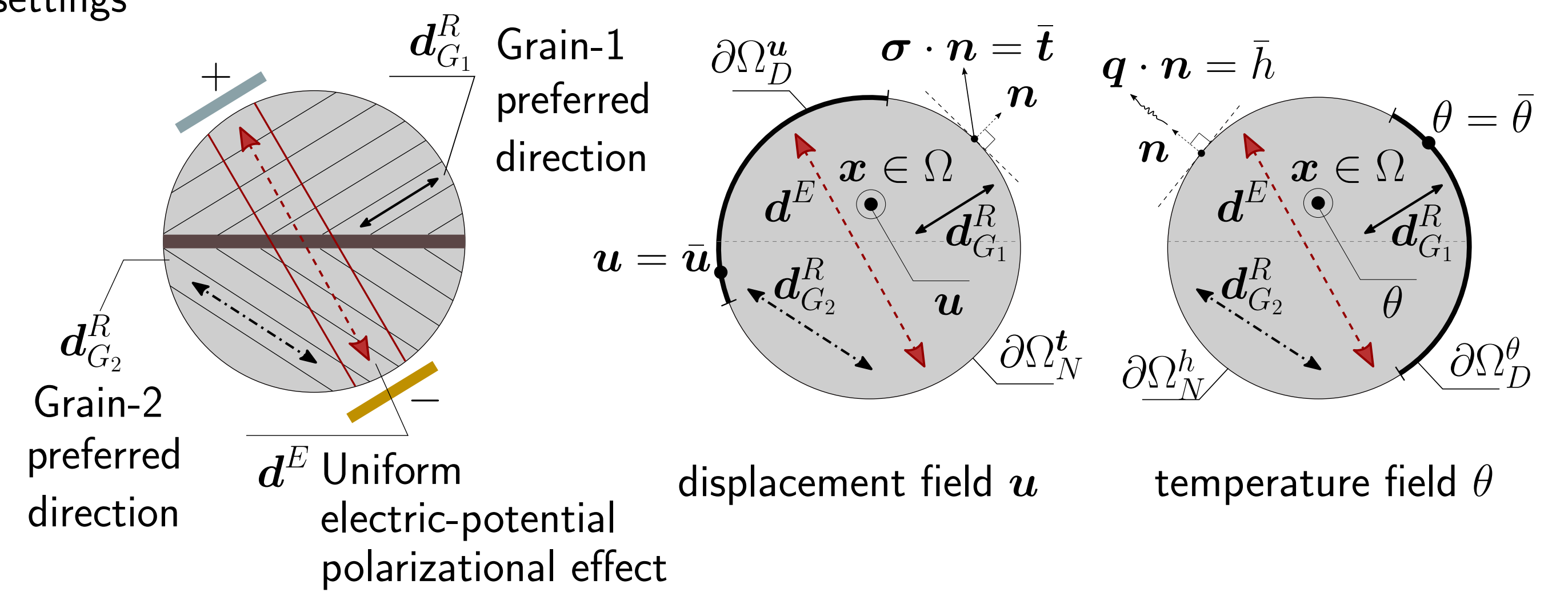
## Nucleation interface: Taking place at the critical dendritic interface

Boundary settings

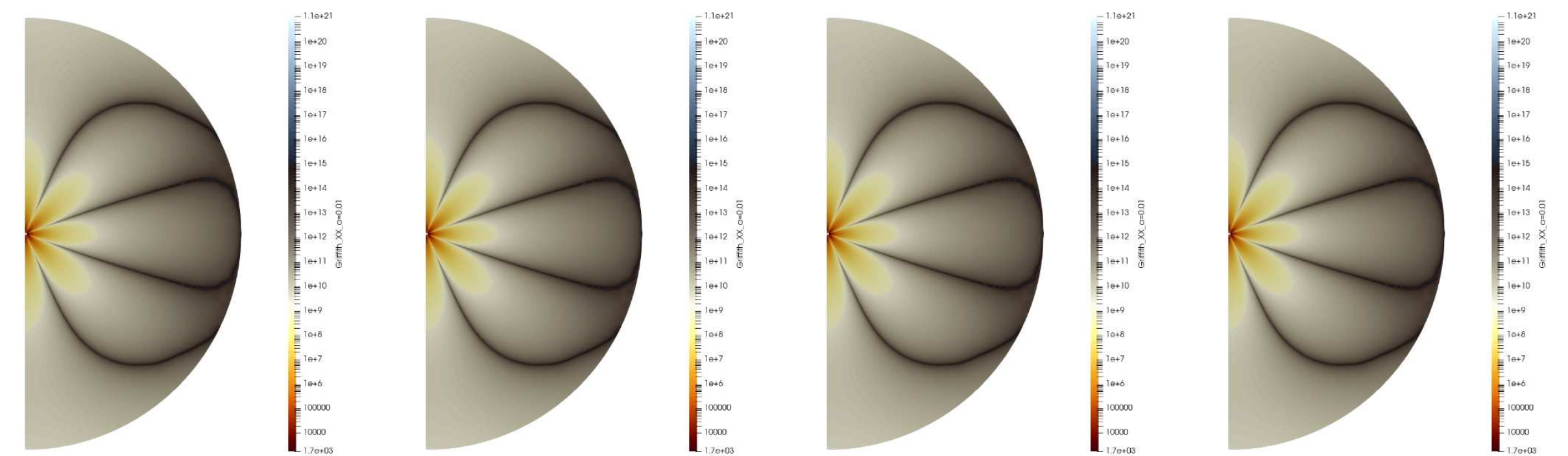
$$\begin{aligned} & \rho \partial_t^2 \mathbf{u}^{(s)} + \nabla \cdot \left( \mathbb{C}^{(D)}_{(\lambda, \mu)} : \nabla \mathbf{u}^{(s)} \right) + \rho \nabla V_e = \mathbf{0}, \\ \text{s.t. } & a_{\text{Griffith}} := a^* = \arg \min_{a \in \mathbb{R}} \iint_{\Omega} f(a, \mathbf{u}; \lambda, \mu, \mathbf{d} \otimes \mathbf{d}) d\Omega - \iint_{\Gamma} f(a; \gamma) d\Gamma \Big|_{\mathbf{u}^{(s)}} \\ & \rho \partial_t^2 \mathbf{u}^{(s)} + \nabla \cdot \left( \mathbb{C}^{(D)}_{(\lambda, \mu)} : \nabla \mathbf{u}^{(s)} \right) + \rho \nabla V_e = \mathbf{0}, \\ \text{s.t. } & a_{\text{Griffith}} := a^* = \arg \min_{a \in \mathbb{R}} \iint_{\Omega} f(a, \mathbf{u}; \lambda, \mu, \mathbf{d} \otimes \mathbf{d}) d\Omega - \iint_{\Gamma} f(a; \gamma) d\Gamma \Big|_{\mathbf{u}^{(s)}} \\ & \rho \partial_t^2 \mathbf{u}^{(s)} + \nabla \cdot \left( \mathbb{C}^{(D)}_{(\lambda, \mu)} : \nabla \mathbf{u}^{(s)} \right) + \rho \nabla V_e = \mathbf{0}, \\ \text{s.t. } & a_{\text{Griffith}} := a^* = \arg \min_{a \in \mathbb{R}} \iint_{\Omega} f(a, \mathbf{u}; \lambda, \mu, \mathbf{d} \otimes \mathbf{d}) d\Omega - \iint_{\Gamma} f(a; \gamma) d\Gamma \Big|_{\mathbf{u}^{(s)}} \\ & \rho \partial_t^2 \mathbf{u}^{(s)} + \nabla \cdot \left( \mathbb{C}^{(D)}_{(\lambda, \mu)} : \nabla \mathbf{u}^{(s)} \right) + \rho \nabla V_e = \mathbf{0}, \\ \text{s.t. } & a_{\text{Griffith}} := a^* = \arg \min_{a \in \mathbb{R}} \iint_{\Omega} f(a, \mathbf{u}; \lambda, \mu, \mathbf{d} \otimes \mathbf{d}) d\Omega - \iint_{\Gamma} f(a; \gamma) d\Gamma \Big|_{\mathbf{u}^{(s)}} \\ & \rho \partial_t^2 \mathbf{u}^{(s)} + \nabla \cdot \left( \mathbb{C}^{(D)}_{(\lambda, \mu)} : \nabla \mathbf{u}^{(s)} \right) + \rho \nabla V_e = \mathbf{0}, \\ \text{s.t. } & a_{\text{Griffith}} := a^* = \arg \min_{a \in \mathbb{R}} \iint_{\Omega} f(a, \mathbf{u}; \lambda, \mu, \mathbf{d} \otimes \mathbf{d}) d\Omega - \iint_{\Gamma} f(a; \gamma) d\Gamma \Big|_{\mathbf{u}^{(s)}} \end{aligned}$$

Therefore

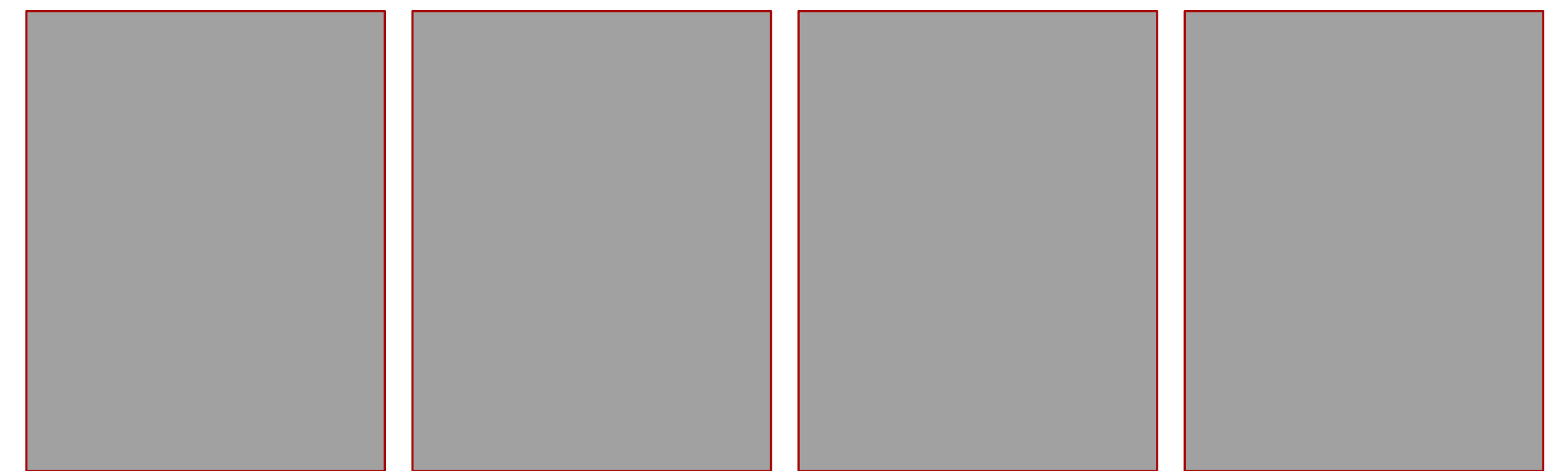
$$\therefore a_{\text{Griffith}} := a^* = \arg \min_{a \in \mathbb{R}} \iint_{\Omega} f(a, \mathbf{u}; \lambda, \mu, \mathbf{d} \otimes \mathbf{d}) d\Omega - \iint_{\Gamma} f(a; \gamma) d\Gamma \Big|_{\mathbf{u}^{(s)}}$$



The set of boundary conditions is likewise the path of the pressure-centric dendritic crack.



Comparison: Analytical vs. Numerical solutions



### Contact

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### References

- [1] **T.Vo**, *Modeling the swelling phenomena of li-ion batt. cells based on a numerical chemo-mech. coupled approach*. MA, Robert Bosch Battery Systems GmbH, 2018.
- [2] **S.Braun**, C.Yada and A.Latz, *Thermodynamically consistent model for Space-Charge-Layer formation in a solid electrolyte*. Jr. Phys. Chem., 119, 22281-22288, 2015.
- [3] **C.Hüter**, S.Fu, M.Finsterbusch, E.Figgemeier, L.Wells, and R.Spatschek, *Electrode-electrolyte interface stability in solid state electrolyte system: influence of coating thickness under varying residual stresses*. AIMS materials Science, 4(4):867-877, 2017.
- [4] **S.Kim**, J.S.Kim, L.Miara, Y.Wang, S.K.Jung, S.Y.Park, Z.Song, H.King, M.Badding, J.M.Chang, V.Roev, G.Yoon, R.Kim, J.H.Kim, K.Yoon, D.Im, and K.Kang, *High-energy and durable li metal batt. using garnet-type solid electrolytes with tailored li-metal compatibility*. Nature Communications, 13(1):1883, 2022.