Strong Localization and Rapid Time Scales of Superheating in Solid-State Nanopores

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Nanopore Heating

- Voltage source (V)
- 70 nm thickness
- 100 nm distance
- 3M NaCl solution
- Si$_3$N$_4$ material

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3M NaCl

Si$_3$N$_4$

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Si$_3$N$_4$
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3M NaCl

Si₃N₄
Experimental Results: Pore Conductance

Conductance [$\mu$S] vs. Time [$\mu$s]

Initial Conductance, 1.15 $\mu$S

V = 4.0 V, 5.0 V, 6.0 V, 7.0 V, 8.22 V
Experimental Results: Pore Conductance

Conductance [µS] vs Time [µs]

8.22 V
Experimental Results: Pore Conductance

- Conductance [μS]
- Time [μs]
- Fall Time ~ 1 ns
- 604 K
- 117 ns
- 8.22V
- 16 ns
Motivating Question

How hot is the pore center?
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– No experimental means of measuring
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– Appeal to COMSOL modeling
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Can heating dynamics explain nonlinear conductivity measured before a nucleation event?
COMSOL Modeling

- Geometry
- Governing Equations: Joule Heating
- Material Properties
- Boundary Conditions
- Results
COMSOL Modeling

- Geometry
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Nanopore Geometry

- 2D Axisymmetry
- External boundary at S is on the order of 200 microns
COMSOL Modeling

- Geometry

- Governing Equations: Joule Heating

- Material Properties

- Boundary Conditions

- Results
Governing Equations: Joule Heating

Heat Equation: \[ \rho C_p \frac{\partial}{\partial t} T = \nabla \cdot [\kappa \nabla T] + Q \]
Governing Equations: Joule Heating

Heat Equation: \[ \rho C_p \frac{\partial}{\partial t} T = \nabla \cdot \left[ \kappa \nabla T \right] + Q \]

Source Term: \[ Q = J_t \cdot E \]
Governing Equations: Joule Heating

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Source Term: \[ Q = J_t \cdot E \]

Continuity Equation: \[ \nabla \cdot J_t = q_i \]

\[ J_t = \sigma E + \varepsilon_0 \varepsilon_r \frac{\partial}{\partial t} E + J_{ex} \]

\[ E = -\nabla V \]
Governing Equations: Joule Heating

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Source Term: \[ Q = J_t \cdot E \]

Continuity Equation: \[ \nabla \cdot J_t = q_i = 0 \]

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COMSOL Modeling

- Geometry
- Governing Equations: Joule Heating
  - Material Properties
- Boundary Conditions
- Results
Material Properties

• Require material data for superheated water
  – Not available in COMSOL
  – Obtained from IAPWS-95 equation of state
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• Amorphous Silicon Nitride thin film
  – Different thermal conductivity than bulk
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• What about electrical conductivity of 3M NaCl solution?
Conductivity 3M NaCl Solution

Joule Heating: 
\[ \sigma(T)E^2 \]

COMSOL Modeling

• Geometry
• Governing Equations: Joule Heating
• Material Properties
  • Boundary Conditions
• Results
Boundary Conditions

\[ n \cdot J = 0 \]

\[ V = 0 \]

Voltage Source

Axisymmetric Boundary
COMSOL Modeling

• Geometry
• Governing Equations: Joule Heating
• Material Properties
• Boundary Conditions

• Results
Nanopore Heating

8.22V pulse applied for 10.4µs
Nanopore Heating

8.22V pulse applied for 10.4µs

Temperature [K]

470K  600K

100 [nm]
Experimental Results: Pore Conductance

- Conductance: [µS]
- Time: [µs]
- Voltage Levels: 8.22 V, 7.0 V, 6.0 V, 5.0 V, 4.0 V
- Initial Conductance: 1.15 [µS]
Experimental Results: Pore Conductance

Initial Conductance, 1.15 μS

Conductance [μS]

Time [μs]

8.22 V 603 K
7.0 V 485 K
6.0 V 409 K
5.0 V 359 K
4.0 V 329 K
Conclusions

• Nanopore heating experiments
  – Temperature at the center of the pore: 600K
  – Close to kinetic limit of superheat
  – Not possible to experimentally measure

• Modeled using COMSOL Joule Heating Module
  – Flexibility to incorporate specialized material data
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Governing Equations: Joule Heating

$$\nabla \cdot \left( \sigma E + \frac{\partial}{\partial t} D \right) = \nabla \cdot (\sigma E) + \frac{\partial \rho}{\partial t} = 0$$
Governing Equations: Joule Heating

\[ \nabla \cdot \left( \sigma \mathbf{E} + \frac{\partial}{\partial t} \mathbf{D} \right) = \nabla \cdot (\sigma \mathbf{E}) + \frac{\partial \rho}{\partial t} = 0 \]

\[ \frac{\partial \rho}{\partial t} = -\nabla \cdot (\sigma \mathbf{E}) = -\nabla \sigma \cdot \mathbf{E} - \sigma \nabla \cdot \mathbf{E} \]
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Governing Equations: Joule Heating

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\[ \frac{\partial \rho}{\partial t} = -\nabla \sigma \cdot \mathbf{E} - \frac{\sigma \rho}{\epsilon} \]

Not zero!