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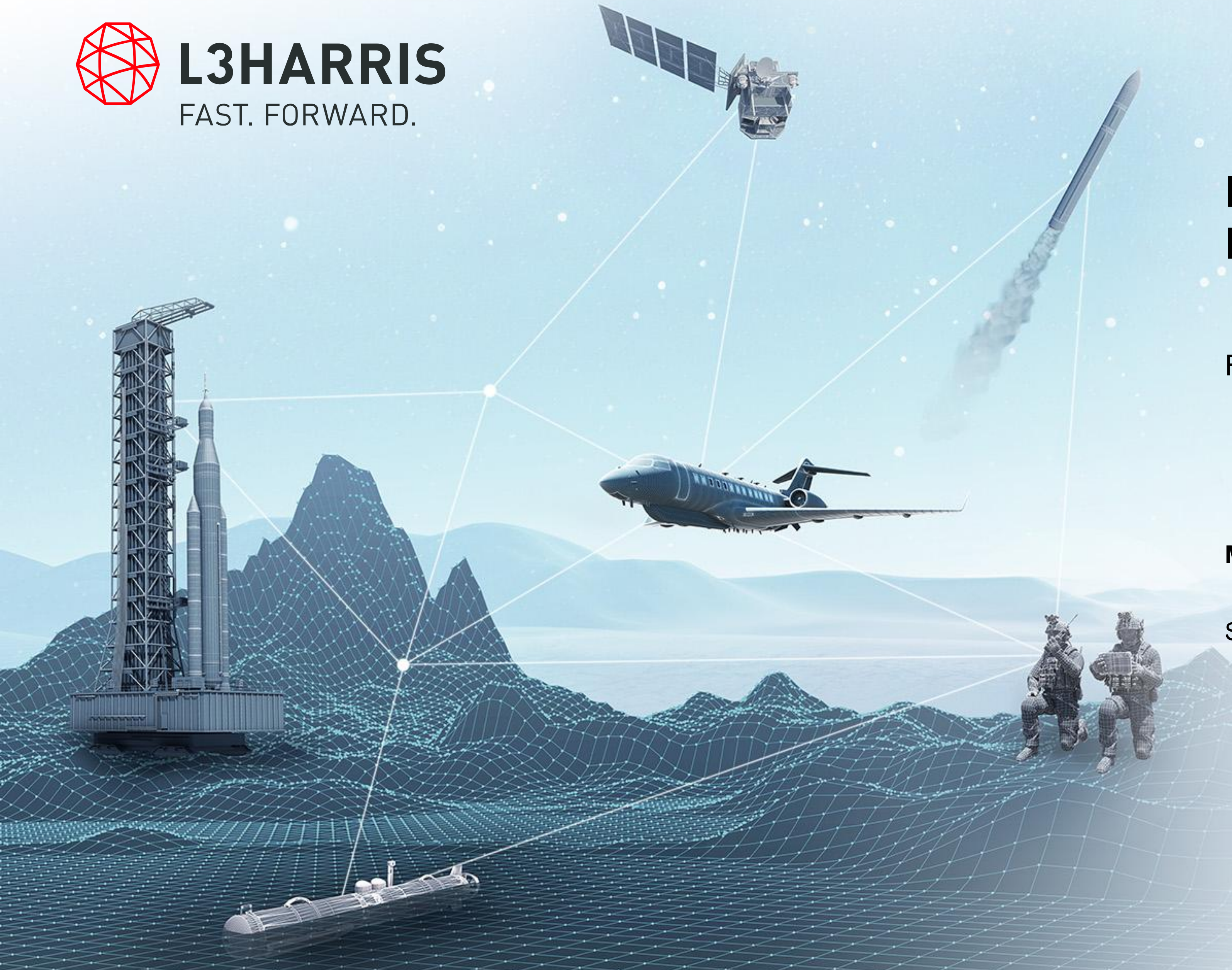

NANODIAMOND THICK FILMS FOR THERMAL MANAGEMENT

Fabrication and characterization

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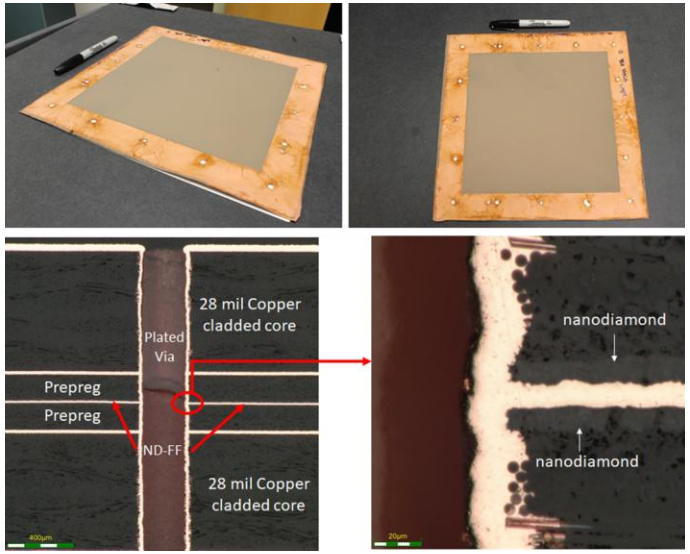
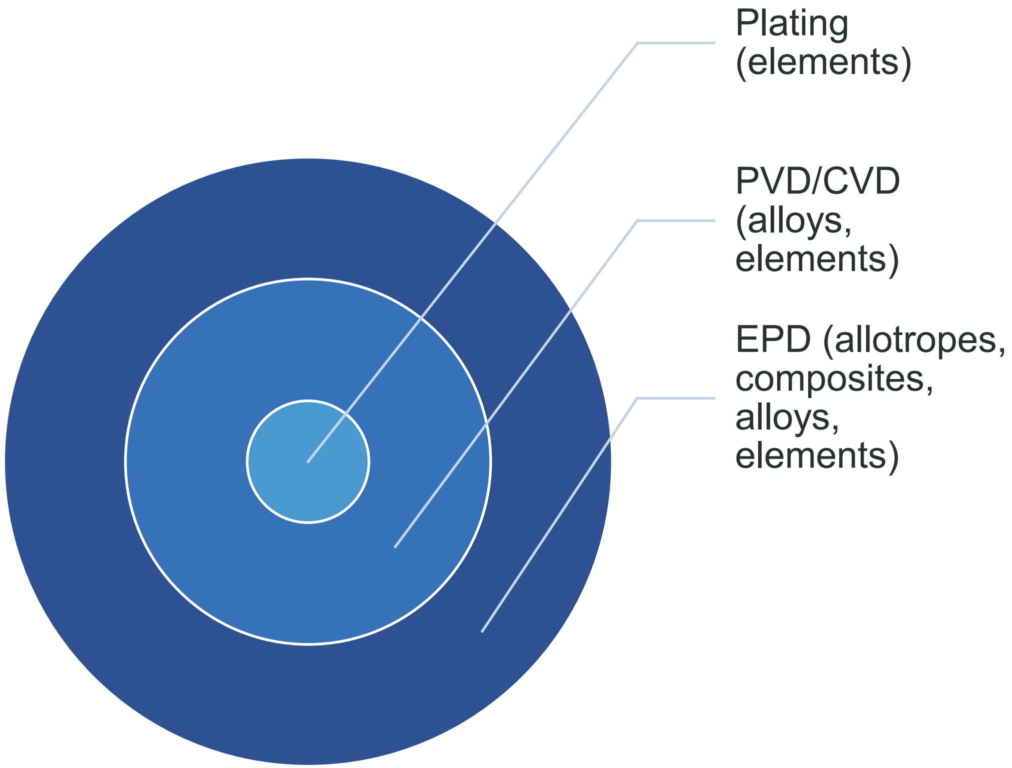
Nanodiamond thick films for thermal management

Problem: The decreasing size of modern electronic devices, combined with increasing performance demands, leads to higher heat density in smaller spaces. Efficient diffusion or extraction of this thermal energy limits the miniaturization of these heat-generating electronics.

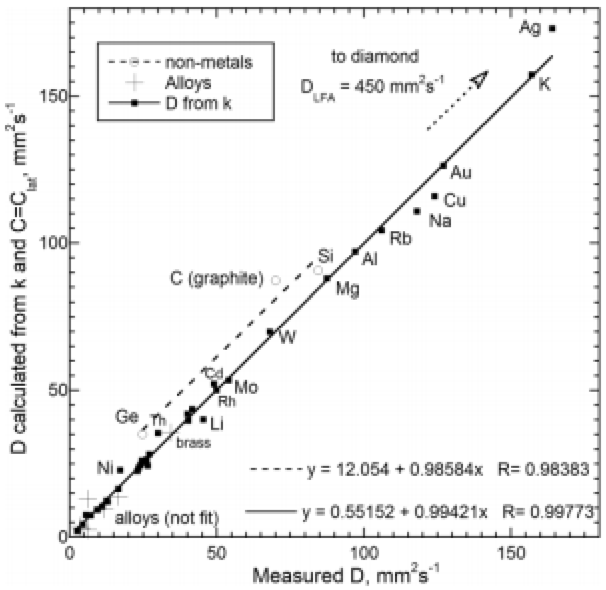
Solution: Electrophoretic deposition of nanodiamond allows for high thermal conductivity (~1,600 W/mK) films at low cost and flexibility of integration.

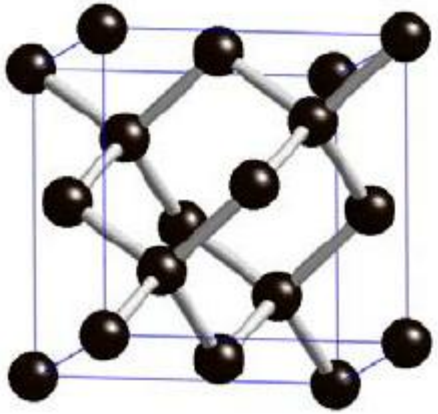




Diamond – A unique thermal conductor

- Mostly high thermal conductivity materials are electrically conducting
  - – Electronic contributions to thermal conductivity are generally high in metals
- However, diamond has a high phonon contribution to conductivity through its tight carbon-carbon bonding, **around 1000-2200 W/(m·K)**, multiple times higher than the highest thermal conductivity metal
- Diamond is much lighter than metallic thermal conductors at **3.51 g/cm³**
- Other useful properties:
  - – Highest hardness of known materials*
  - – Low dielectric loss tangent 0.00005 at 145 GHz
  - – Dielectric constant of around 5.7
  - – Breakdown strength of ~100 MV/m





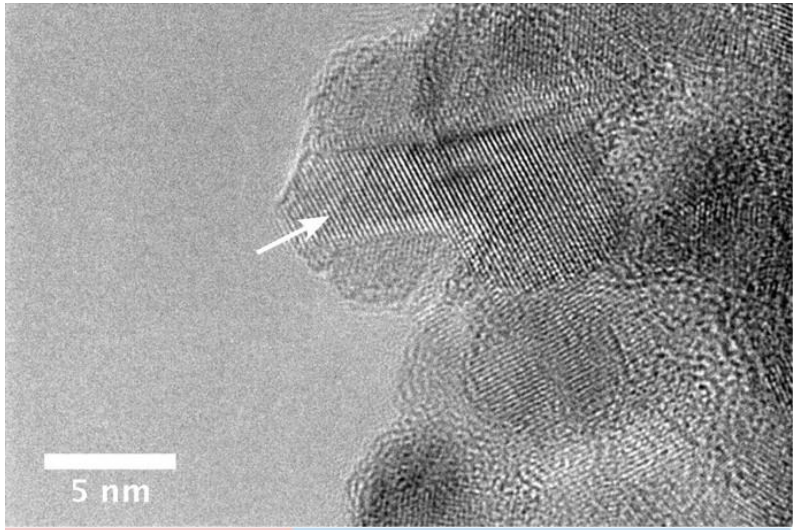
Above: D, a measure of phonon contribution to thermal conductivity

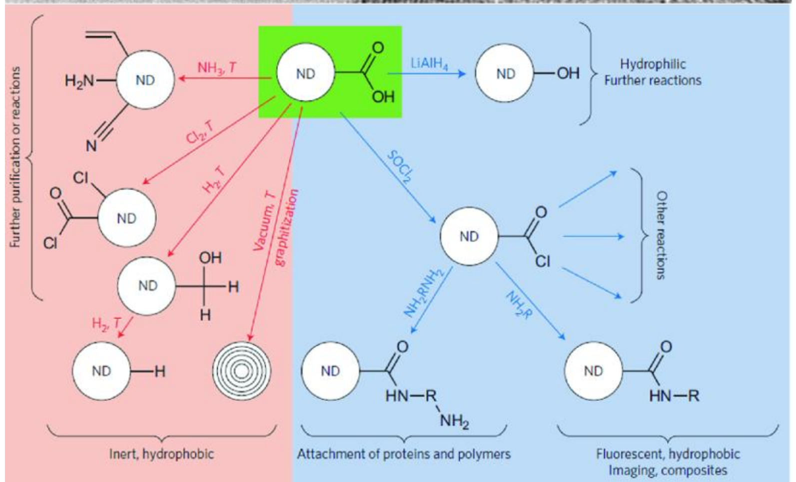
Left: Diamond lattice

Criss, E. M., & Hofmeister, A. M. (2017). Isolating lattice from electronic contributions in thermal transport measurements of metals and alloys above ambient temperature and an adiabatic model. *International Journal of Modern Physics B*, *31*(14), 1750205.

Detonation nanodiamond

- Detonation nanodiamond are nanoscale diamond particles formed by the detonation of carbon containing explosives
- The resultant carbon mixture with some metal contaminants is then purified with fuming nitric acid followed by a furnace treatment in atmosphere
- Nanodiamonds formed by this method are ~5 nm, largely with surface oxygen and hydroxyl groups

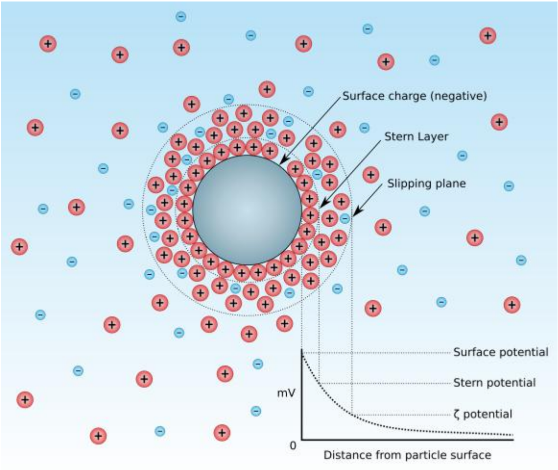


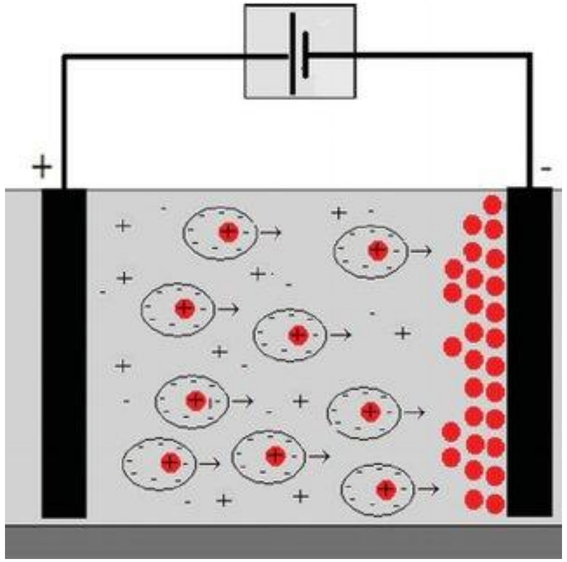


Top: Afandi, A., Howkins, A., Boyd, I.W. *et al.* Nanodiamonds for device applications: An investigation of the properties of boron-doped detonation nanodiamonds. *Sci Rep* **8,** 3270 (2018). https://doi.org/10.1038/s41598-018-21670-w
Bottom: Ashek-I-Ahmed, Perevedentseva E.V., Karmenyan A., Cheng CL. (2019) Spectroscopy of Nanodiamond Surface: Investigation and Applications. In: Yang N. (eds) Novel Aspects of Diamond. Topics in Applied Physics, vol 121. Springer, Cham

Electrophoretic deposition

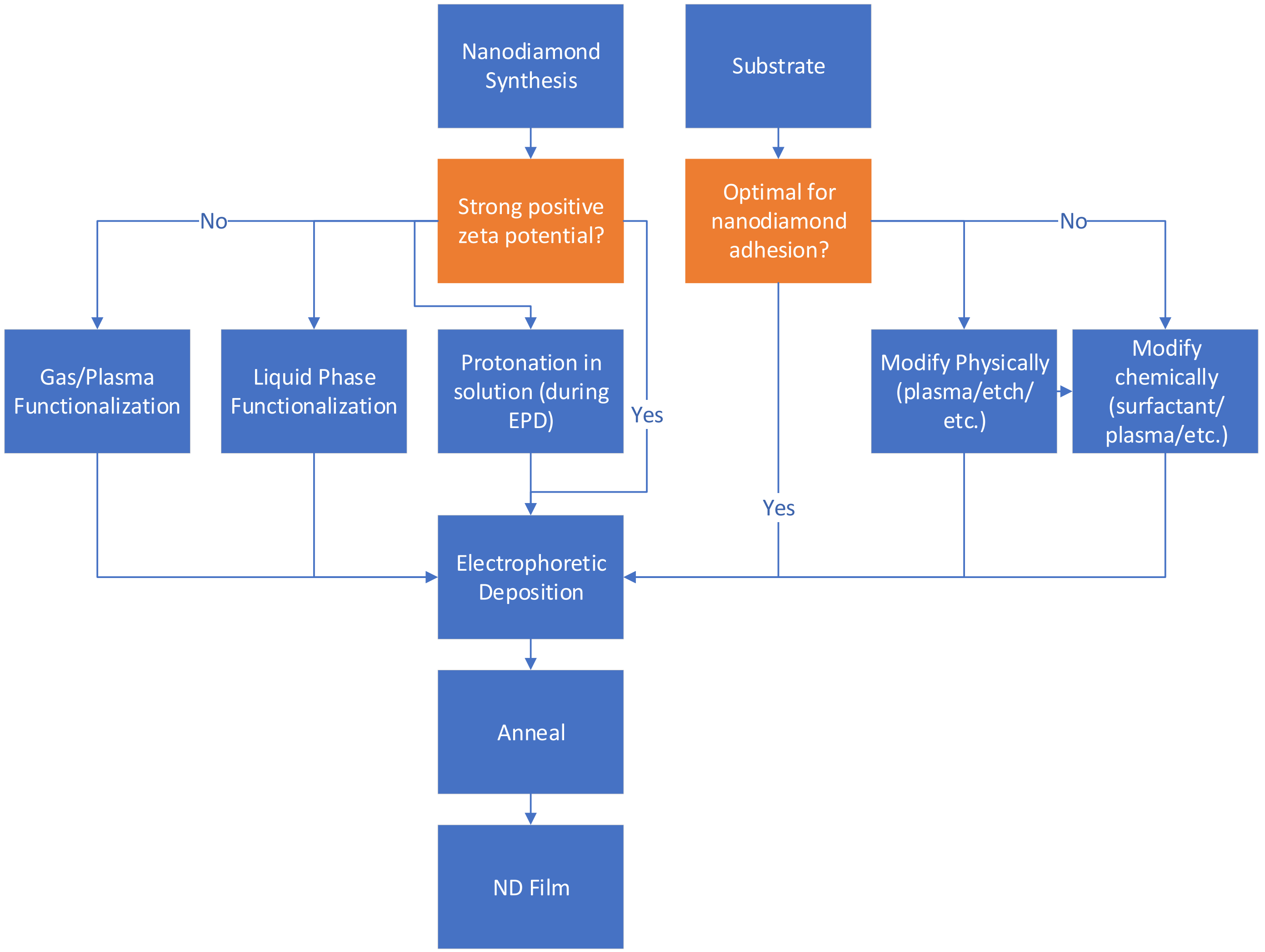
- In electrophoretic deposition, particles are suspended in solution tuned such that their surface becomes charged
- Then, the particles are exposed to an electric field to induce movement on the charged particles
- It is important to have a high magnitude zeta potential (top figure) to prevent agglomeration of particles in solution





Top image: https://commons.wikimedia.org/wiki/File:Diagram_of_zeta_potential_and_slipping_planeV2.svg
Bottom image: Ammam, Malika. (2012). Electrophoretic deposition under modulated electric fields: A review. RSC Advances. 2. 7633-7646. 10.1039/C2RA01342H.

Process flow



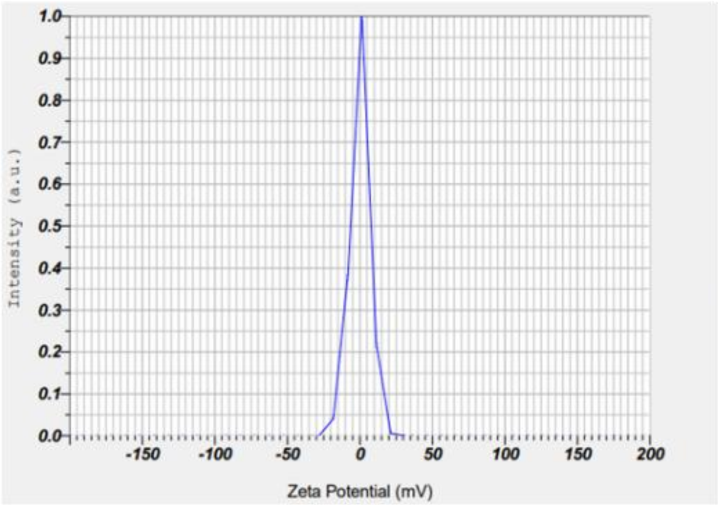
Generating a strong positive zeta potential

- We sought a strong positive zeta potential to prevent agglomeration with graphene and promote cathodic deposition
- We could not reach a strong positive zeta potential by adjusting pH alone, so we used iodine and acetone to generate protons to protonate the nanodiamond surface

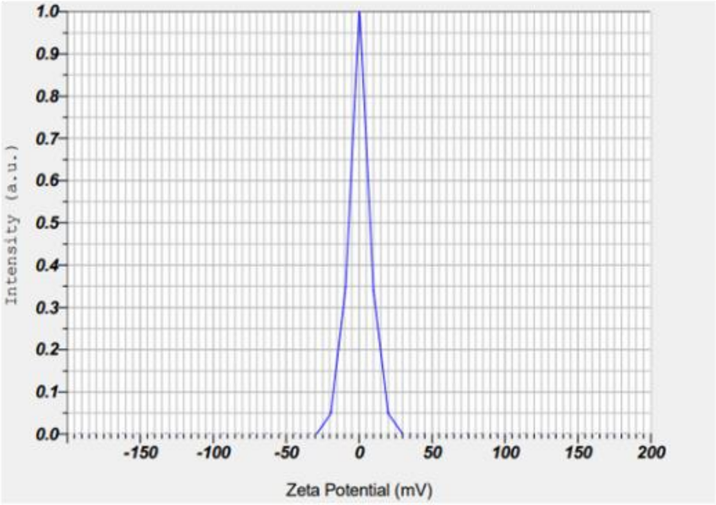
$$CH_3 - CO - CH_3 + 2I_2 \Leftrightarrow ICH_2 - CO - CH_2I + 2H^+ + 2I^-$$

- Other types of surface modifications may be more suitable to different systems and goals

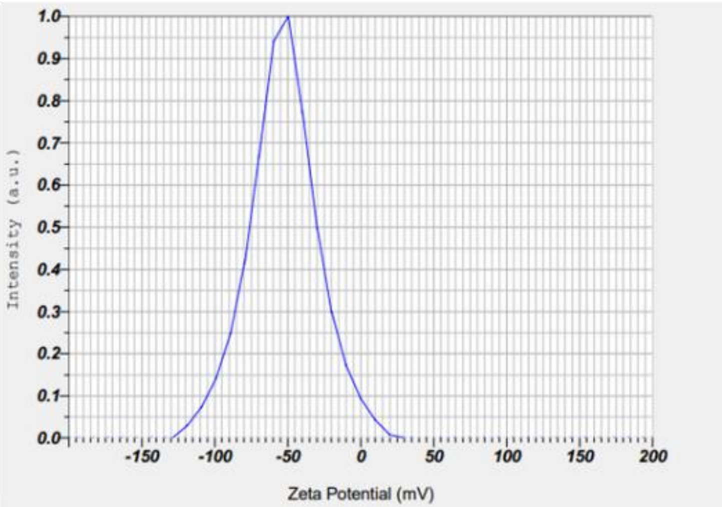
pH = 4



pH = 7

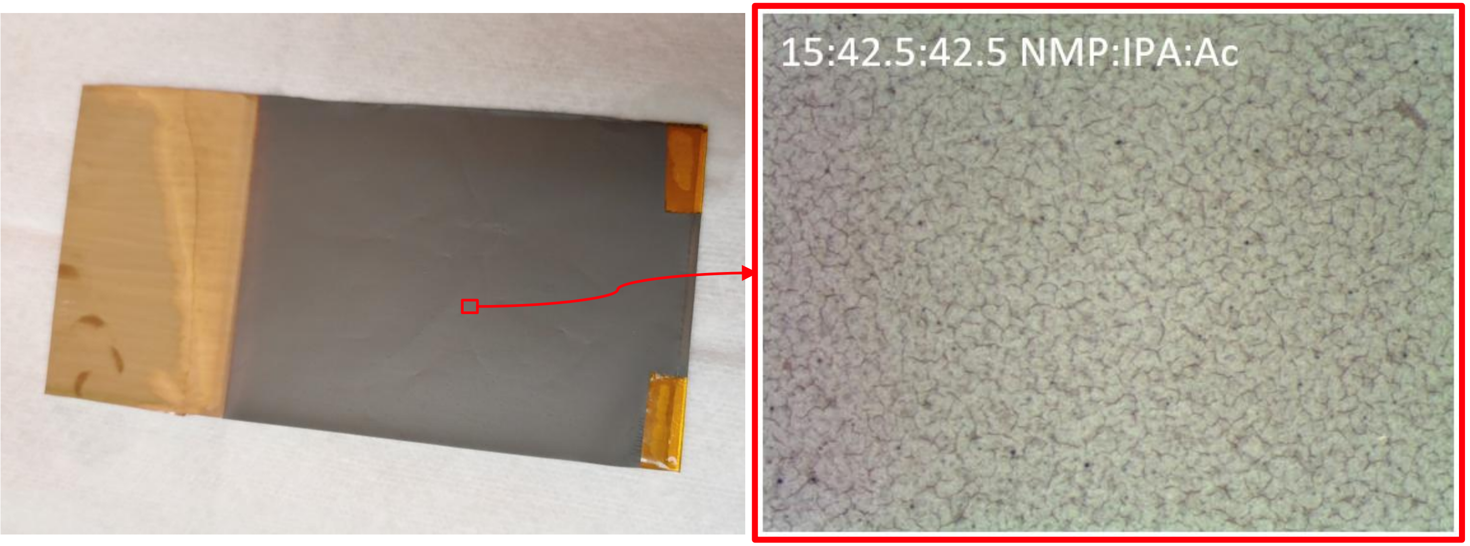


pH = 10



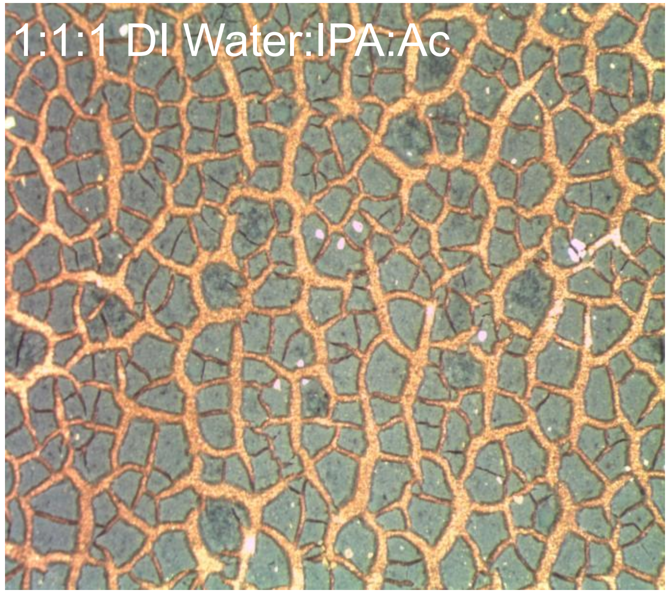
Mitigating cracking in thick films

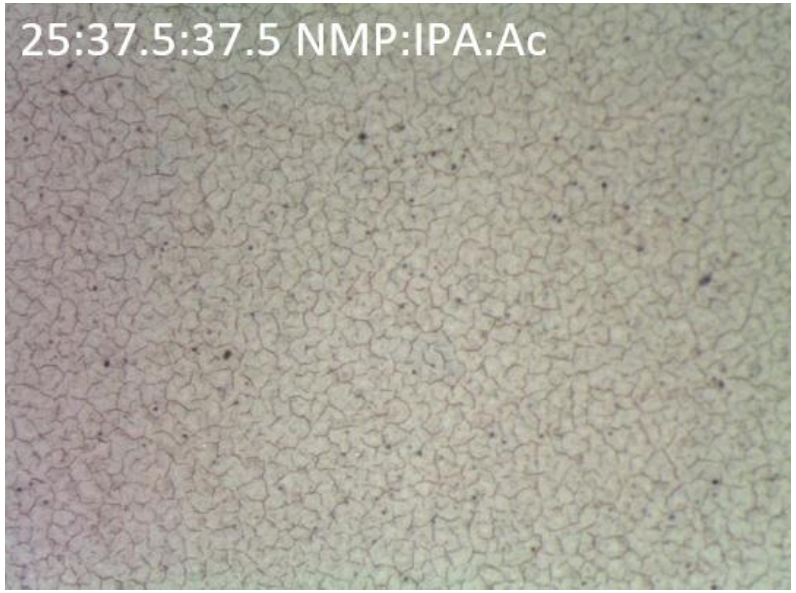
- Different surface treatments were attempted – micro-etch, oxidation, various plasma, etc.
- Critical factors to surface preparation:
  - Roughness
  - Surface tension
  - Attractive short-range forces to nanoparticles

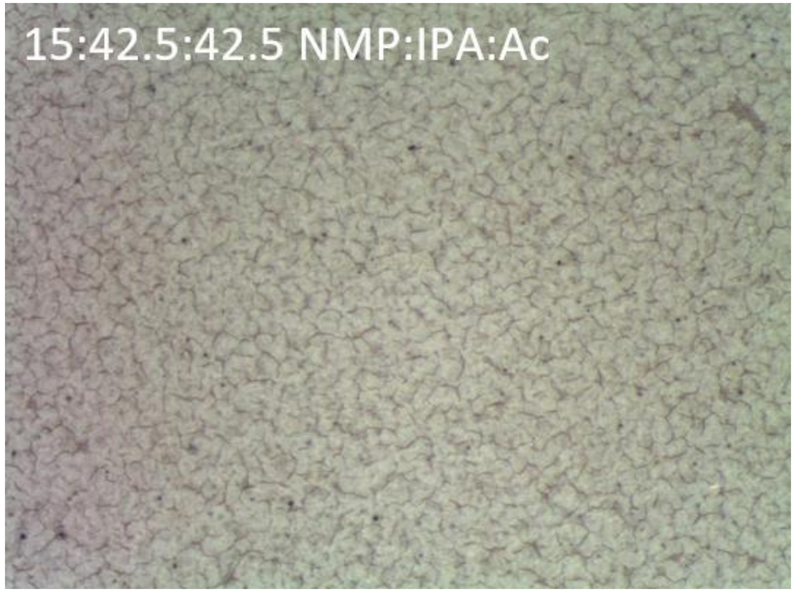


Solution properties

- Solution properties which need to be tuned for optimal deposition:
  - Sufficiently high permittivity: To allow for high zeta potential, for dispersion and electromotive force
  - Low surface tension: Wettability of substrate and decrease in capillary forces
  - Low evaporation rate: Lower capillary forces and more economic bath maintenance
- Tuned by solvent selection and ratio







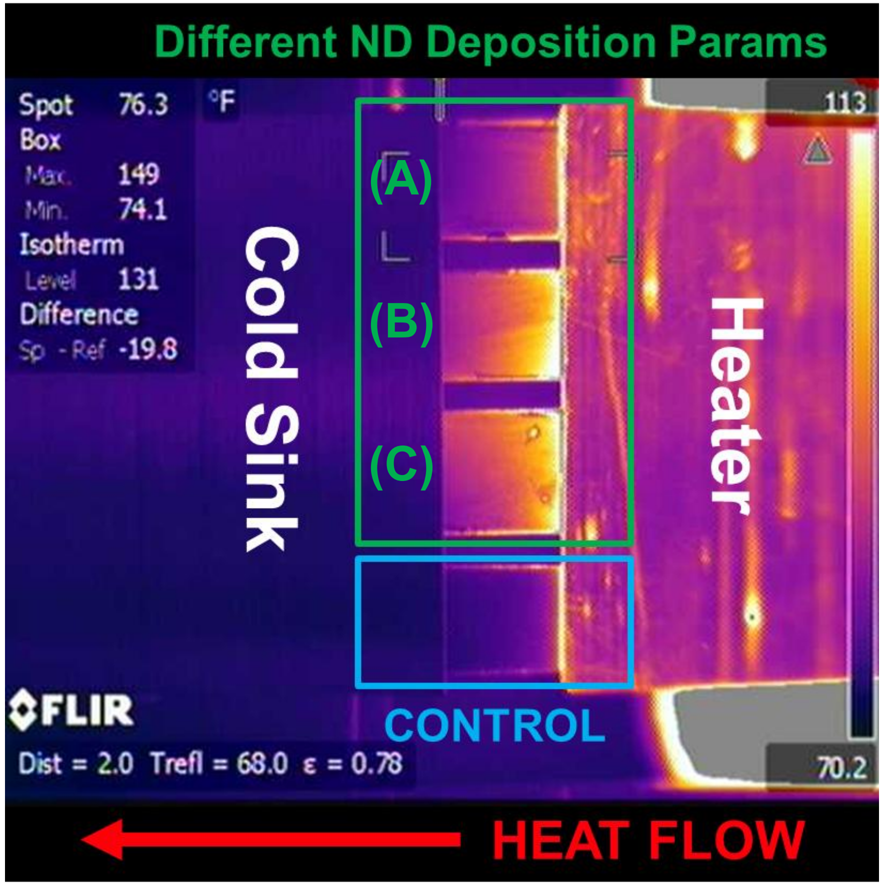
Enhanced thermal conductivity demonstration

Increased Conductivity

- Nanodiamond films showed a drastic increase in thermal transfer from the heater to the cold sink compared to the copper control

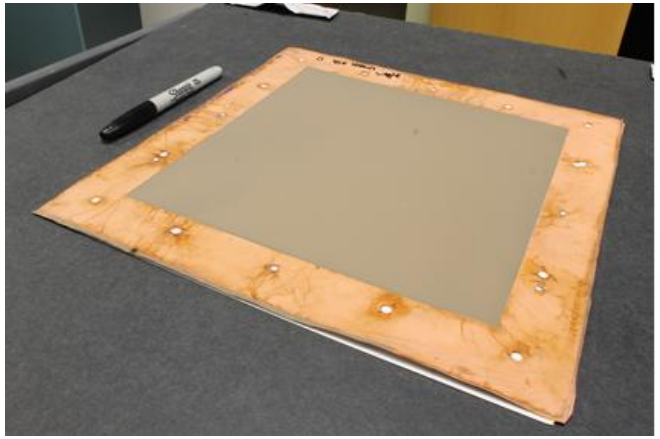
Critical Demonstration

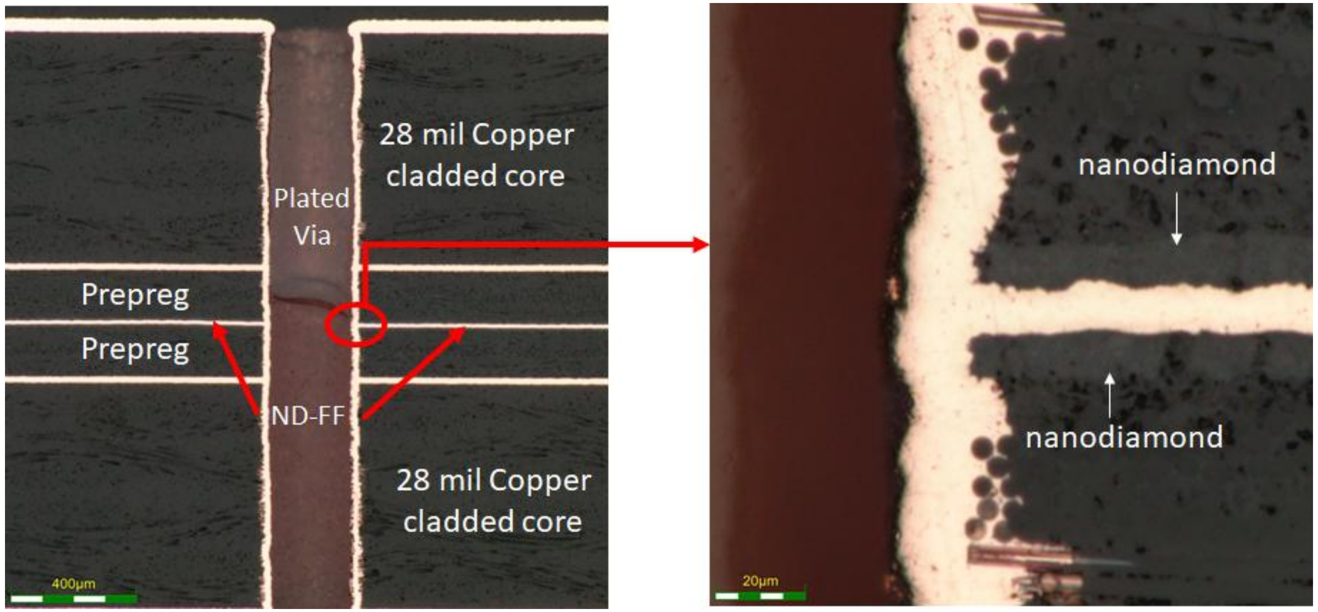
- Achieved sufficiently dense films
- Nanodiamonds exhibit similar thermal conductivity to bulk diamond
- Optimized deposition parameters for nanodiamond density/conductivity



Large area format coatings and lamination

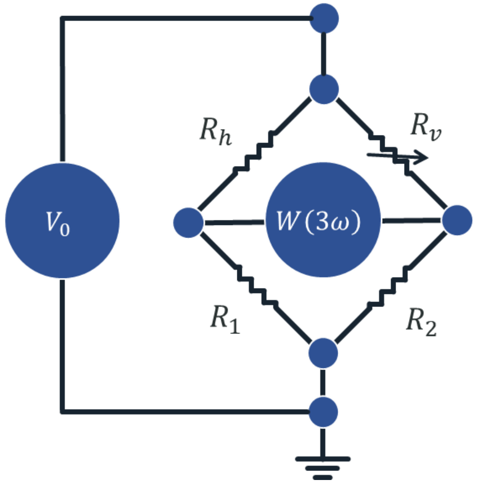
- Large area copper foil substrates were coated with nanodiamonds and laminated into multi-layer laminates with high adhesion
- Multiple lamination runs were performed to demonstrate robustness of the process

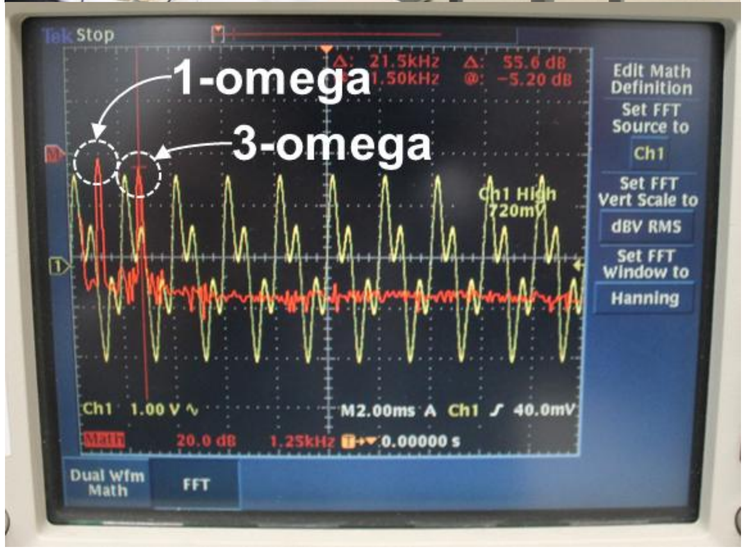




3-Omega thermal measurement: Theory

- Principle:
  - Voltage/Current at 1-omega
  - Heats substrate at 2-omega (absolute value of current)
  - Substrate heats and cools at 3-omega changing resistance
  - Resistance change measured shows cooling rate
  - Calculate thermal conductivity
- Instrumentation:
  - Wheatstone bridge provided cleanest signal of available approaches, although it's less flexible than an instrumental amplifier scheme which allows for quicker setup for 1 omega signal removal
  - Reduce noise and temperature fluctuations with insulated faraday cage
- Observations:
  - It took much more time to achieve temperature stability from the RMS voltage with lower thermal conductivity materials
  - Temperature stability reached nearly instantaneously with nanodiamond sample





3-Omega thermal measurement: Results

Solving for thermal conductivity, k, in

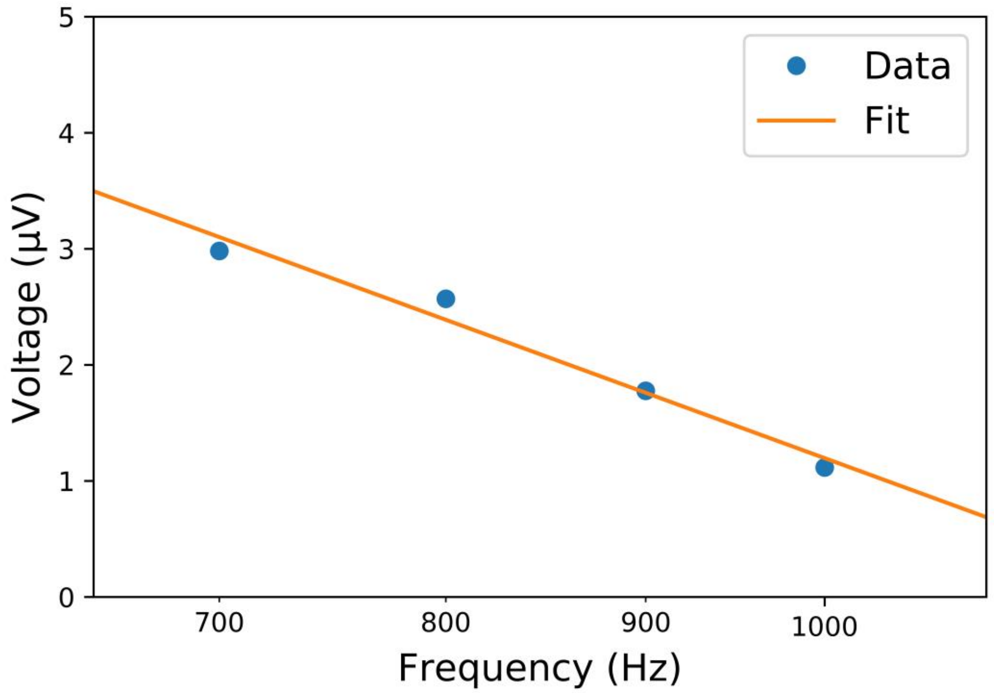
$$W(3\omega) = slope \cdot \ln(2\omega) + intercept$$

$$slope = V^3 \frac{beta}{4pi \cdot l \cdot k \cdot R_h} * \left(\frac{R_1}{R_h + R_1}\right)$$

Where l is the length of the heater, R_h is the heater resistance, R_1 is the resistor in series with Rh in the wheatstone bridge, $beta$ is the TCR of the copper trace, V is the applied voltage at 1ω, and $W(3\omega)$ is the measured voltage across the wheatstone bridge at 3ω.

- Slope = -5.34e-06, Intercept = 4.18e-05, Stderr = 6.18e-07
- Note: the stderr in the slope fit is low which is good, although propogation of errors is not straightforward in this case. Also, the 0.08 W/mK for the subtrate was not considered but this should have minimal effect due to it being several orders of magnitude lower
- Solving for k gives 1639 W/(mK)

The thermal conductivity of the nanodiamond film was measured to be 1600 W/(mK), ~73% of bulk diamond and approximately 400% higher than copper or silver



Conclusions and outlook

Nanodiamond for Thermal

- Nanodiamond films have shown promise for high thermal conductivity thermal solutions
- Further improvements can be made to enhance film density and further mitigate cracking

Nanodiamond for Electrical

- For dielectric applications, validation of complete elimination of cracking and pinholes will be required
- Additional uniformity metrics will need to be met

Other Material Systems

- While nanodiamond thick films are novel, electrophoretic deposition is a widely published microfabrication method, although not prevalent in microelectronics outside academia
- Various electrophoretically deposited films may be relevant to microelectronic systems

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