Atomic Whispers to System Health Diagnosis and Prognosis: First-Principles-Based Degradation Modeling of 2D Materials in Next-Generation Bioelectronics

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ABSTRACT

Our results demonstrate that Cr intercalation into 2D transition metal dichalcogenide materials will significantly elevate the interlayer shear resistance, acting as an atomic-scale "glue" that mitigates delamination and structural failure—a key degradation mechanism. We uncover dynamic stabilization mechanisms and quantify the energy barriers that retard lateral sliding, which are crucial inputs for physics-of-failure models. We combine *ab initio* density functional theory (DFT) and machine-learned-force-field Molecular Dynamics (MLFF-MD) for this study. Leveraging MLFFs allows us to extend our simulations to larger length- and time-scales and hence capture long-term dopant dynamics and degradation evolution. MLFF-MD has the advantage of combining a bigger scope with near-DFT accuracy, enhancing predictive capabilities for materials design.

This work provides mechanistic insight into transition metal intercalation for 2D material's stabilization and offers a physics-informed computational framework for assessing material longevity and reliability. Such predictive capabilities are critical for proactive Prognostics and Health Management (PHM), enabling the rational design of robust 2D heterostructures, guiding synthesis strategies, and informing maintenance protocols for advanced electronic and spintronic devices.

1. PROBLEM STATEMENT

Low-dimensional and so-called van der Waals (vdW) materials(Geim & Grigorieva, ,) are driving the development of next-generation electronic and bioelectronic systems that could enable miniaturized, stretchable, and transient devices essential for applications like implantable healthcare. These devices promise real-time diagnosis, personalized treatment, and functional restoration. However, ensuring their long-term reliable operation and effective Prognostics and Health Management (PHM) is a significant challenge, especially because of the need to address consideration of the complex degradation pathways inherent to these materials, coupled with the scarcity of reliability data, create a critical "blind spot" in the advancement of PHM. There is an urgent need for reliability prediction tailored specifically to the unique requirements of implantable and biocompatible devices.

Conventional PHM models, often rooted in macroscopic continuum mechanics or purely data-driven approaches, fundamentally fail to capture the degradation processes occurring at the atomic scale in atom-thin 2D materials. This calls for a new paradigm and framework for PHM. The core PHM challenge for these materials can be articulated as this query: How to achieve understanding of atomic-scale degradation, accelerate application involving this understanding to realistic scales and timescales via machine learning, translate this into long-term lifetime prediction, and ultimately provide actionable engineering guidance?

Antimony Telluride (Sb_2Te_3), a layered chalcogenide, exemplifies the promise of these materials for thermoelectric and sensing applications(Venkatasubramanian, Siivola, Colpitts, & O'Quinn,), particularly when doped with Cr for added functionalities like ferromagnetism.(Chang et al.,)However, achieving precise doping and reliable property measurements, especially in thin-film form, presents significant experimental challenges, often involving tedious and time-consuming lab- oratory procedures. Despite preliminary experimental findings showing enhanced properties(Wang et al.,), a comprehensive understanding of 2D material's degradation mechanisms and long-term reliability under operational stresses (thermal, mechanical, electrical, environmental) remains largely unexplored(Novoselov, Mishchenko, Carvalho, & Castro Neto,). Traditional experimental approaches to systematically study these degradation pathways across var-

ious conditions are often impractical. Fortunately, computational capabilities like density functional theory (DFT) and molecular dynamics (MD) simulations, offer powerful tools for the rational design of materials and the prediction of their properties.(Zeni et al., ,). Dopant configurations, defects, interfaces, and environment significantly influence stability, but these factors are poorly characterized experimentally. This knowledge gap, exacerbated by experimental limitations, prevents the development of robust, physics-informed PHM strategies. Addressing these vulnerabilities requires a proactive investigation that bridges the atomic and macroscopic scales.

This research aims to bridge this critical knowledge gap by establishing the necessary physics-informed framework. Employing a multiscale computational approach,(Momeni et al.,) we use high-fidelity first-principles DFT for fundamental interactions and defect energetics, leveraging this data to train Machine Learned Force Fields (MLFFs)(Unke et al.,). MLFFs enable accelerated MD simulations to track degradation over long timescales under realistic stresses, incorporating physics-informed principles. This framework provides the scientific basis for physics-informed PHM, with multiple levels of impact: enabling early "Design for Reliability," improving forecasting for implantable systems, and linking atomic degradation signals to macroscopic health indicators for potential real-time monitoring.

2. EXPECTED NOVEL CONTRIBUTIONS TO PHM

This research advances PHM for 2D vdW materials in bioelectronics through our multiscale computational framework. The key contributions are:

1. **Atomic-Scale Degradation Understanding**: Establish physics-based degradation mechanisms using DFT and MLFF-MD to identify failure precursors and quantify Physics-of-Failure (PoF) parameters. This approach surpasses continuum methods by revealing atomic-level mechanisms that govern electronic, magnetic, and photonic properties, enabling deeper understanding of material performance in biological environments and PoF modeling for new materials lacking experimental data.

2. **Hybrid Prognostic Model improve Predictability**: Combine atomistic simulation insights with machine learning to create a hybrid model driven by physical parameters to forecast degradation trends.

3. **Proactive Reliability Framework**: Demonstrate a validated DFT→MLFF-MD→Hybrid ML workflow for early weakness identification that enables reliability-informed design and real-time health assessment. This framework extends to other 2D materials, bioelectronics, and thermoelectric systems.

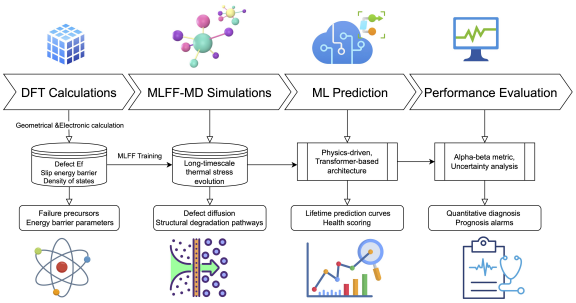

Figure 1. Flowchart of the Multiscale Workflow

These advances bridge atomic-scale physics and practical PHM for next-generation bioelectronics.

3. RESEARCH PLAN

3.1. Overall Approach

This research establishes a comprehensive multiscale computational workflow to bridge the gap between atomistic material behavior and macroscopic PHM outputs for doped vdW materials like Cr-doped Sb_2Te_3. The methodology progresses from fundamental atomic-scale calculations to accelerated dynamics simulations and ultimately to a physics-informed prognostic model, providing actionable insights for reliability engineering. The overall research pipeline, illustrating the flow between modules, is conceptually shown in Figure 1.

The first phase leverages DFT calculations to understand defect formation, preferred doping sites, and migration pathways. This provides essential insights into potential failure mechanisms at the atomic scale.

We then develop Machine Learned Force Fields (MLFFs) based on DFT data, enabling efficient simulation of larger systems. These force fields help explore how defects and strain affect material properties.

Using these tools, we can simulate material degradation under realistic conditions, considering factors like temperature, mechanical stress, and defect interactions. The results directly inform our predictive models for material reliability.

By combining DFT precision with MLFF-accelerated MD efficiency, this multiscale approach effectively simulates vdW material degradation under operational conditions (temperature, strain, doping, defects). The resulting data—including defect evolution and structural changes—directly inform the Physics-Informed Prognostic Model, enabling accurate and efficient reliability predictions.

3.2. Work Performed & Preliminary Results

Initial computational investigations have been conducted to establish baseline properties of Cr-doped Sb_2Te_3 (Figure 2a-c) and explore fundamental aspects critical for understand-

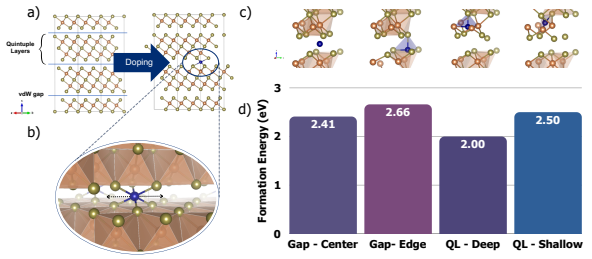

Figure 2. **Structural configuration and doping strategy of Cr-doped Sb_2Te_3.** (a) Schematic illustration of the Cr doping process into the Sb_2Te_3 layered structure, highlighting the van der Waals (vdW) gap and quintuple layers (QLs). (b) Enlarged view of the doped structure, showing the Cr atom positioned within the vdW gap and its coordination with neighboring atoms. (c) Atomic visualizations of various Cr doping sites: Gap-Center, Gap-Edge, QL-Deep, and QL-Shallow. (d) Calculated formation energies for each doping configuration, indicating that QL-Deep doping is the most energetically favorable with a formation energy of 2.00 eV.

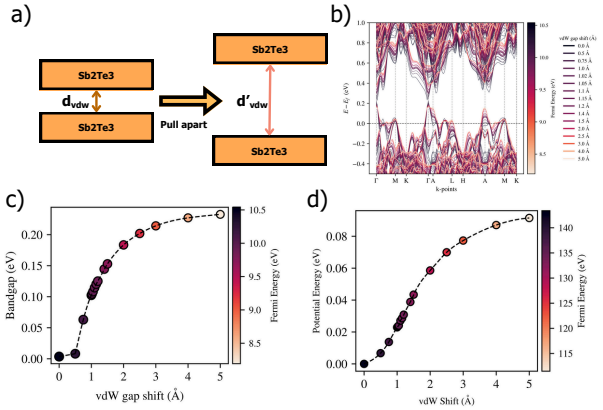

Figure 3. **Band gap modulation and energetics as a function of van der Waals (vdW) gap expansion in Sb_2Te_3.** (a) Schematic illustration showing the widening of the vdW gap between adjacent quintuple layers. (b) Calculated band structures for Sb_2Te_3 with natural ($\Delta d = 0$ Å) and expanded ($\Delta d = 5$ Å) vdW gap, revealing a pronounced increase in band gap with increasing interlayer separation. (c) Band gap value as a function of vdW gap distance, showing systematic widening up to $\Delta 0.25$ eV as the gap expands. (d) Potential energy change versus vdW gap distance, demonstrating that moderate separation (up to 5 Å) requires minimal energy input (< 0.08 eV).

ing its degradation, thereby demonstrating the feasibility of the proposed multiscale approach and providing foundational data for the physics-informed prognostic model.

Characterization of the Cr Dopant Site: We made DFT calculations of formation energy (E_f) corresponding to different Cr dopant positions in the Sb_2Te_3 crystal. Results indicate that an interstitial Cr buried within the quintuple layer is thermodynamically preferred, showing lower E∗f and reduced strain (Figure 2d). This helps identify the likely initial Cr dopant locations for studying degradation mechanisms.

vdW Gap Engineering for Band Structure Tuning: We investigated the impact of incrementally widening the vdW interlayer distance using DFT (Figure 3a). This study demonstrated that widening the vdW gap leads to a smooth and reversible modulation of the electronic band gap (Figure 3b-c). This validates our hypothesis that the vdW gap distance can, in principle, serve as an effective "control knob" for "band structure engineering" in Sb_2Te_3-based systems if wider vdW gaps can be achieved in practice. Understanding the relationship between the vdW gap and electronic properties is vital, as degradation processes (like delamination or intercalation) can alter this gap, directly impacting device functionality.

Room Temperature Interface Stabilization via AIMD: *Ab initio* Molecular Dynamics (AIMD) simulations were performed on small supercells to estimate atomic-scale properties at realistic temperatures, in contrast to traditional (0K) DFT calculations. Our AIMD calculations show that Cr interstitials suppress thermal fluctuations in the vdW gap, playing a dampening role in interfacial dynamics. This suggests that interstitial Cr may contribute to stabilizing the layered structure against thermal stresses, providing initial insights into Cr's potential role in mitigating certain degradation pathways.

Cr as an Atomic Glue – Migration Barrier Analysis: Nudged Elastic Band (NEB) calculations were employed to calculate energy barriers along prescribed reaction coordinates, revealing insights into atomic migration (Figure 4a). Our NEB calculations reveal that Cr doping introduces a 1 eV increase in the in-plane sliding barrier between quintuple layers of Sb_2Te_3 (Figure 4b). This significant enhancement in resistance to lateral shear suggests that Cr dopants tend to function as interlayer stabilizers, effectively "gluing" adjacent layers together. This quantitative increase in the migration barrier is a critical input for parameterizing kinetic models of structural degradation, indicating enhanced mechanical stability due to Cr doping.

Electronic Properties Preserved Post-Doping: PDOS and band structure analyses show semiconducting behavior is maintained in Cr-doped Sb_2Te_3 (1-2 Cr atoms/120-atom system), with a direct, narrow band gap (Figure 3b) crucial for thermoelectric properties. Higher Cr concentrations induce unwanted metallic behavior, indicating that an optimal doping window exists that balances stability and electronic properties while preventing degradation-induced metallic transitions.

These preliminary results confirm the feasibility of the computational approach and lay the groundwork for future work.

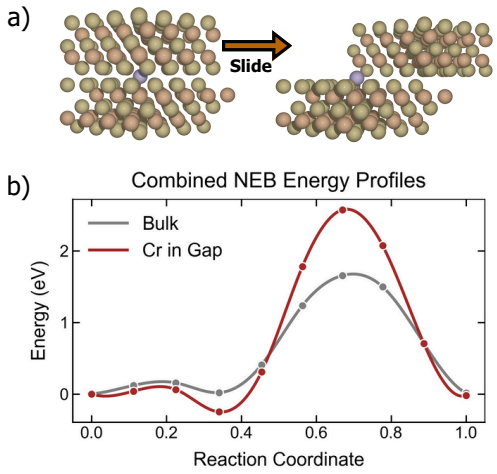

Figure 4. **Effect of Cr intercalation on interlayer dislocation and shear resistance in Sb_2Te_3.** (a) Schematic visualization depicting the in-plane dislocation of the upper quintuple layer (QL) relative to the lower QL, used to simulate interlayer sliding under shear stress. (b) Calculated Nudged Elastic Band (NEB) energy profiles for layer displacement in pristine (black) and Cr-intercalated (red) Sb_2Te_3. Cr incorporation markedly increases the activation barrier for sliding from 1.65 eV to 2.57 eV, indicating enhanced resistance to mechanical dislocation.

3.3. Future Work

Building on the preliminary findings and the multiscale computational framework, future research is likely to focus on the following key areas:

1. **Investigating Coupled Degradation via Accelerated MD:** Utilize suitably trained MLFFs for large-scale, long-timescale MD simulations investigating degradation under coupled thermal, environmental, and mechanical stresses, including heterostructure stability and dynamic phenomena.
2. **Mapping Microscopic to Macroscopic Performance:** Construct quantitative mapping functions between simulated atomic-scale defects and macroscopic property degradation, integrating atomic insights into macroscopic models for reliability assessment.
3. **Uncertainty Quantification and Experimental Validation:** Quantify simulation uncertainties and rigorously cross-validate computational predictions against accelerated experimental aging data, ensuring model robustness and predictive accuracy across more materials and scenarios.

4. CONCLUSION

The reliable operation of next-generation electronic and thermoelectric devices, particularly those based on advanced 2D vdW materials like Cr-doped Sb_2Te_3 for applications including implantable bioelectronics, is critically dependent on a thorough understanding of their intrinsic stability and degradation pathways. This research proposes a comprehensive investigation utilizing a multiscale computational framework, integrating high-fidelity first-principles DFT with efficient MLFF-accelerated MD simulations, to uncover the fundamental atomistic mechanisms governing the degradation of Cr-doped Sb_2Te_3 under various operational stressors.

The goal of this Ph.D. research is to provide the foundational scientific understanding and predictive tools necessary for developing effective, physics-informed PHM strategies. By elucidating atomistic degradation mechanisms and generating crucial quantitative parameters, this work will directly inform and enable more accurate and reliable prognostic models. The advantage of the proposed approach lies in its ability to combine atomistic precision from DFT with the large-scale, long-timescale simulation capabilities of MLFF-accelerated MD, thereby bridging the gap between fundamental material physics and macroscopic reliability prediction. Ultimately, this research aims to enhance the prognostic capabilities for devices employing 2D vdW materials, facilitating their confident and reliable deployment in future advanced electronic and sensing technologies.

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