EXTENDED ABSTRACT

This dissertation, entitled *Predicting and Analyzing the Thermal and Electrical Properties of Materials Using Advanced Machine Learning Models*, develops correction and prediction frameworks for thermal conductivity (TC) and electrical conductivity (EC) characterization in thin films using a combination of experimental methodology, physics-based modelling, and advanced machine learning (ML). The work is divided into two major components. The first addresses TC determination using Scanning Thermal Microscopy (SThM) and focuses on reducing roughness-induced artifacts by integrating simultaneous thermal–topographical mapping, normalization strategies, and ML regression. The second addresses EC determination using Four-Point Probe (4PP) measurements and focuses on mitigating geometry-induced distortions using both Finite Element Method (FEM) simulations and a data-driven ML correction framework trained solely on experimental measurements.

The thesis is guided by two primary hypotheses. The first hypothesis states that integrating SThM thermal mapping with detailed surface topography analysis, together with ML models trained on physically meaningful descriptors, can overcome the inherent limitations of SThM analysis and yield accurate and reproducible thin-film TC estimates. The second hypothesis states that both FEM and ML can generate effective correction factors for 4PP conductivity measurements by compensating for geometric distortions such as edge effects and probe placement sensitivity. While FEM is expected to provide high-fidelity physics-based corrections, ML is hypothesized to offer a scalable and simulation-free alternative capable of comparable accuracy when trained on representative experimental datasets.

1. Thermal conductivity: motivation, limitations of existing methods, and the SThM challenge

Thermal conductivity κ is one of the most fundamental thermophysical properties, describing how efficiently a material transfers heat under a temperature gradient. In submicron-scale devices, TC measurements are extremely affected by surface topography. TC characterization is typically performed using optical pump–probe techniques such as Time-Domain Thermoreflectance (TDTR) and Frequency-Domain Thermoreflectance (FDTR). These methods offer accurate non-contact characterization, can probe layered systems and buried interfaces, and have been extensively validated in the literature. In TDTR, heat is deposited in a thin metallic transducer and the cooling response is monitored through reflectivity changes. TC is extracted by fitting measured time-dependent cooling curves to theoretical diffusion models, typically interpreted via lock-in detected in-phase/out-of-phase signals. Despite their accuracy and versatility, TDTR/FDTR suffer from inherent spatial resolution limitations due to the diffraction limit of light, restricting conductivity maps to the micron scale. As a result, TDTR/FDTR cannot reliably capture submicron-scale spatial TC variations at sample surfaces.

In contrast, Scanning Thermal Microscopy (SThM) provides submicron lateral resolution by integrating AFM scanning with a thermal probe. SThM enables simultaneous acquisition of topography and thermal signals, which is a powerful advantage for local thermal characterization in complex systems.

However, quantitative TC extraction using SThM remains challenging. The measured thermal response is strongly affected by probe–sample interaction conditions, including surface roughness and mechanical contact variability. Even small submicron changes in topography can disturb the tip–sample contact, distort heat flux paths, and produce thermal artifacts. Surface asperities reduce true contact area, creating localized air gaps that increase thermal contact resistance (TCR) and alter the effective thermal resistance sensed by the probe. The result is that SThM thermal maps often represent a complex convolution of intrinsic TC, topography-dependent contact effects, and substrate contributions (especially in thin films with thickness below 100 nm). Mechanical contact models such as DMT theory, although foundational, encounter limitations when applied to irregular morphologies and asymmetric contact geometries typical of real thin-film surfaces.

This thesis identifies that the key obstacle is lack of a reliable and scalable correction approach that can decouple intrinsic TC from topography-induced measurement artifacts. Therefore, the thermal component of this dissertation is designed to build an integrated methodology that combines SThM thermal mapping, correlated topographical characterization, normalization strategies, and ML regression.

2. Thermal methodology: normalization, topography descriptors, and ML framework

2.1. Normalization using quartz reference and definition of Γ_i

One of the central methodological approach in this thesis is the implementation of a quartz-referenced normalization procedure to reduce probe-dependent variability and enhance comparability across samples. Amorphous quartz is selected as a reference material due to its thermal stability and isotropic heat conduction properties, enabling reliable baseline calibration. The SThM signal is processed by comparing the difference between dynamic and static electrical resistances of the probe measured on the sample and on the quartz reference at corresponding spatial coordinates. This yields a dimensionless normalized thermal signal ratio:

$$\Gamma_i = \frac{R_d^{s_i} - R_s^{s_i}}{R_d^{n_i} - R_s^{n_i}}.$$

This normalized ratio cancels probe-specific and instrumental influences and captures the effective thermal interaction between the probe and the sample relative to a stable reference. R_d and R_s and are dynamic and static electrical resistances of the probe, respectively. Γ_i is a physically meaningful parameter. This is quantitatively supported in the thesis through correlation analysis: the raw resistance difference shows negligible predictive relevance, while the normalized Γ_i becomes the strongest contributor to TC prediction.

2.2. Multi-scale mapping strategy and dataset structure

The experimental framework applies a multi-scale spatial mapping methodology. A $2 \times 2\ \mu\text{m}^2$ region of each sample is partitioned into a 16×16 microgrid (256 cells). This grid-based approach provides spatial robustness against localized anomalies and generates a dataset appropriate for ML training. Within this region, localized submicron windows of $375 \times 375\ \text{nm}^2$are analyzed and divided into 3×3 submicron grids. The methodology is designed to establish a spatially correlated dataset linking local thermal response to corresponding local topography. Both trace and retrace maps are collected and averaged to reduce noise and improve reliability.

The thesis emphasizes that the aim is not simply to map thermal contrast but to generate a numerical dataset that translates real surface topography into quantitative descriptors that can be used as ML inputs. This approach converts a metrology limitation into a structured data problem.

2.3. Topographical descriptors at micro and submicron scales

The framework incorporates micro-scale topographical descriptors based on statistical surface metrology, primarily Root Mean Square roughness R_{rms}^s and surface skewness R_{sk}^s. RMS roughness reflects the amplitude of surface irregularity and influences effective contact area. Skewness describes the asymmetry of height distribution and distinguishes peak-dominated vs valley-dominated surfaces, each imposing different thermal transport constraints.

At the submicron scale, localized descriptors are extracted from 3×3 grids, including surface inclination M^{s_i}, peak-to-valley variation μ^{s_i}, and local height distribution variability. These descriptors explicitly represent local geometric effects that strongly influence tip–sample contact mechanics. In the thesis, these submicron parameters are presented as essential contributors to explaining variability in the measured thermal resistance response.

2.4. Substrate–thickness factor *C*

A key innovation in the thesis is the development of a substrate–thickness factor C as an ML input to distinguish bulk from thin-film behavior and incorporate substrate influence. This distinction is crucial because thin films exhibit fundamentally different thermal transport regimes, and substrate contributions dominate especially below a threshold thickness comparable to the SThM interaction length scale ($\sim$ 100 nm). The thesis defines C as a piecewise function:

$$C = \begin{cases} \left(\frac{b-d}{b}\right)\frac{\kappa_{\text{sub}}}{\kappa^{n}}, & d < b \\ 0, & d \geq b \end{cases}$$

where d is film thickness, b is the threshold thickness, κ_{sub} is substrate TC, and κ^{n} is quartz TC used for normalization. This formulation mathematically encodes the shift from layered substrate-dominated transport to bulk-like transport.

3. Thermal ML target definition and feature relevance

Instead of directly predicting TC as a single value from local measurements (which can be unstable due to contact variability), the thesis defines a continuous physical target variable:

$$\chi = \frac{\kappa^{s}}{\sigma^{s_i}}.$$

Here, σ^{s_i} is the standard deviation of surface heights within a localized submicron cell. This choice transforms TC prediction into a continuous regression task with improved generalization capability, enabling the model to interpolate between values and learn meaningful patterns from multidimensional features.

Feature selection is performed using Spearman's rank correlation coefficient. The thesis reports that Γ_i exhibits the strongest inverse correlation with $\chi(r_s = -0.726)$, confirming that normalized thermal interaction is the dominant predictor. Submicron topographical descriptors such as $\mu^{s_i}(-0.609)$ and $M^{s_i}(-0.572)$ also show strong negative relationships, reinforcing the thesis conclusion that roughness-induced effects are not secondary disturbances but dominant drivers of measurement distortion. The substrate–thickness factor C also shows meaningful negative association (-0.518). In contrast, the phase difference $\Delta\varphi_i$ shows negligible correlation (-0.022) and thus contributes minimally to predictive capability under the studied conditions.

This quantitative relevance analysis supports the core thesis claim that the measured TC cannot be reliably extracted without explicitly accounting for surface morphology and substrate regime effects.

4. Thermal ML model development and results

A dataset of 3,332 thermal measurements is used in the analysis, including 2,352 measurements for model development and cross-validation and 980 for independent validation. The dataset includes annealed ITO thin films deposited on glass substrates and ZnO thin films deposited on silicon substrates, along with bulk reference materials

spanning a wide TC range from 0.17 to 450 $\mathrm{W\,m^{-1}K^{-1}}$. Thermal measurements are performed using AFM-based SThM (Park Systems XE-70) with calibrated probe currents and lock-in amplification for signal stability. Two probe types are discussed to support reproducibility, and all experiments are conducted in a vibration-damped chamber at 25°C. The thesis compares Random Forest regression and Gradient Boosting regression models. Hyperparameter tuning is performed through extensive grid search combined with cross-validation, resulting in the evaluation of 279 distinct models. The Random Forest approach consistently outperforms Gradient Boosting in predictive accuracy and generalization, and the best Random Forest model is selected as Model 'a'. The reported predictive performance of Model 'a' demonstrates high reliability, with strong R^2 and low RMSE values. A final model trained on combined cross-validation data is validated against an independent test set of 980 unseen measurements, confirming generalization.
Using the predicted χ, TC values are derived and compared against known reference values for 17 materials (thin films and bulk). The predicted TC values demonstrate strong agreement across several orders of magnitude. Low and moderate TC materials exhibit particularly high precision, with near-perfect agreement for glass and YAG and excellent accuracy for PMMA. High TC systems such as SiC and bulk ZnO show slightly larger deviations but remain within acceptable ranges, consistent with the thesis observation that model sensitivity decreases at extreme χ values due to training dataset distribution.

5. Electrical conductivity: motivation, limitations, and correction need

Accurately determining EC in thin films is complicated by geometric distortions, particularly edge effects, which distort current distribution and invalidate ideal theoretical assumptions. Two widely used techniques are the van der Pauw method (vdP) and the 4PP technique. vdP provides reliable sheet resistance for homogeneous isotropic films but requires strict conditions such as uniform thickness, negligible contact size, and contact placement along boundaries. It is also time-consuming and effectively destructive due to contact fabrication requirements. Therefore, while vdP is used as a reference technique in this thesis, it is not ideal for routine scalable characterization.
The 4PP method is simpler and more versatile but strongly sensitive to sample geometry, probe spacing, and measurement position. Without corrections, measured EC can deviate significantly from intrinsic conductivity. Therefore, a reliable correction framework is required.

6. Electrical methodology: experimental framework, FEM correction, and ML correction

6.1. Experimental dataset and measurement protocol

Electrical measurements are performed using a dedicated 4PP setup built during the PhD work. Keithley current sources and a nanovoltmeter are used to measure voltage response under controlled current injection. The dataset contains 553 measurements under varied conditions including sample dimensions, probe positions, and sample rotations. The samples include irregular quadrilateral ITO thin films deposited on glass substrates and metallic samples (Cu, W, Ni, Fe, Sn). For ITO samples, measurements are performed at multiple probe positions A_1–A_4 and repeated after systematic counterclockwise sample rotations, ensuring that multiple edge-effect regimes are captured. For metallic samples, measurements are performed once at defined probe positions without rotation.

The ML input variables include sample dimensions L_1, L_2, L_3, L_4, diagonal $L_{\mathrm{diag.}}$, probe position coordinates L_h, L_v, and applied current I.

6.2. FEM correction framework

To validate and correct geometric effects, a 3D FEM model is developed in COMSOL Multiphysics. The simulations reproduce the experimental geometry: a thin conducting sample on a dielectric substrate embedded in air. Electric potential distributions are generated for different probe placements, visually demonstrating how current distortion increases near edges. FEM produces a correction factor based on the relationship between simulated apparent conductivity σ_{num} and intrinsic conductivity σ_a, yielding FEM-corrected conductivity:

$$\sigma_{\mathrm{FEM}} = \left(\frac{\sigma_a}{\sigma_{\mathrm{num}}}\right)\sigma_{\mathrm{exp}}.$$

The thesis shows that σ_{FEM} consistently approaches σ_a, validating the physical correction methodology.

6.3. ML correction framework and definition of Θ

To develop a purely experimental correction approach, the thesis introduces a ML formulation that defines a continuous target variable Θ, enabling regression learning despite limited distinct EC values. Using the ideal analytical relationship between current, voltage, and conductivity in a rectangular 4PP geometry, the thesis defines the ML model as:

$$F_{\mathrm{ML}}(L_1, L_2, L_3, L_4, L_{\mathrm{diag.}}, L_h, L_v, I) = \Theta,$$

and then calculates ML-derived conductivity through:

$$\sigma_{\mathrm{ML}} = \frac{\ln(2)}{2\pi V d}\,\Theta.$$

This formulation transforms the problem from a discrete conductivity class prediction into a continuous physics-informed regression task.

Spearman correlation analysis for the electrical dataset confirms that current I has the strongest monotonic relationship with $\Theta (r_s = 0.96)$, consistent with the formulation.

Random Forest regression is selected and optimized through evaluation of 170 hyperparameter combinations. The best model (Model 'a') is identified using cross-validation metrics.

7. Electrical results: FEM vs ML performance and EC prediction

The thesis compares FEM-corrected results and ML-derived results for both metallic and ITO samples using Relative Difference percentage (RD%) analysis and residual distributions. Both approaches demonstrate strong performance under challenging measurement conditions intentionally focused on edge regions where artifacts are strongest.

For metallic samples, FEM shows a mean RD% of $+5.66\%$ while ML shows -3.15%, with similar variability. This indicates that both methods yield comparably reliable results but with opposite bias directions. For ITO samples, FEM shows mean RD% of -4.30% and ML shows -2.93%, with ML demonstrating tighter error distributions.

Final EC comparisons across nine samples show that ML predictions remain within approximately ±15%of reference values and produce particularly consistent performance across ITO samples. FEM results also demonstrate strong agreement, though variability can increase depending on geometry and boundary conditions.

The electrical component confirms the hypothesis that both FEM and ML can effectively mitigate geometry-induced errors in 4PP measurements. FEM provides physics-based correction with high interpretability, while ML demonstrates strong potential as a scalable simulation-free correction framework when trained on sufficiently representative datasets.