Decoding thermal transport: advanced theories and limits in lattice thermal conductivity

A decades-long theoretical quest actively shaping modern materials theory

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The transport of thermal energy through condensed matter stands as one of the most pervasive and defining phenomena across both fundamental physics and transformative technologies. At the macroscopic scale, the ability to control, direct, or arrest the flow of heat dictates the efficiency of next-generation thermoelectric energy harvesters, governs the thermal management and architectural limits of nanoscale microelectronics, and influences the structural integrity of materials operating under extreme thermodynamic conditions. Yet, beneath these diverse engineering challenges lies a foundational and unifying scientific picture, namely: the full description of how microscopic heat carriers (i.e. collective atomic vibrations or, equivalently, phonons) interact and diffuse through complex crystalline and disordered lattices. Understanding lattice thermal conductivity is not merely a matter of parameter prediction; it is an epistemological and cultural quest within solid-state physics that has spanned decades, continuously reshaping modern materials theory and pushing the boundaries of non-equilibrium statistical mechanics.

This talk is structured to offer a comprehensive conceptual overview of this decades-long theoretical journey, explicitly illuminating the intricate paradigm shifts that occur when moving from fundamental thermodynamics to predictive atomistic simulations. Rather than focusing on specific empirical breakthroughs or material-specific data, the discussion will pivot entirely on the grand theoretical and computational challenges that define the field: namely, the foundational conceptual framing, the formal mathematical development, and the ultimate numerical implementation of heat transport models in condensed phases.

I will begin by establishing the canonical framing of the problem through the elegant lens of Onsager linear-response theory, examining how coupled heat, mass, and charge fluxes collapse into pure thermal conductivity under well-defined physical assumptions.

From this baseline, the seminar will extensively dissect the first major theoretical pillar, namely the phonon Boltzmann Transport Equation (BTE). I will thoroughly explore the transition from the widely adopted Single-Mode Approximation (SMA) —or Relaxation Time Approximation (RTA), which assumes individual phonons thermalize independently to an equilibrium background— to exact, full solutions of the linearized BTE. This includes a deep dive into the iterative and variational formulations (utilizing conjugate gradient methods) required to capture the true non-equilibrium phonon population. Special emphasis will be placed on how quantum perturbation theories, typically implemented via Density Functional Perturbation Theory (DFPT), grapple with the extraction of second- and N-order interatomic force constants to rigorously account for N-phonon scattering rates, mass disorder, and anharmonic perturbations.

Next, the seminar will pivot to the second major pillar, exploring the real-time atomistic picture provided by the Green-Kubo formalism.
Here, the presentation will emphasize how this framework tackles the transport problem directly through the statistical evaluation of the microscopic thermal current. Crucially, by operating entirely in the time domain, the Green-Kubo approach completely bypasses the daunting necessity of resolving the full phonon spectrum along with its inherent harmonic and anharmonic challenges. Instead, the thermal conductivity is extracted from the dynamical fluctuations of the heat flux at thermodynamic equilibrium. The discussion will then confront the

very demanding computational challenges that shift from the formal equations to the numerical implementation: specifically, the stringent requirements for exceptionally long simulation trajectories to achieve statistical convergence of the flux auto-correlation function, alongside the necessity for massive simulation cells to mitigate finite-size effects and properly capture long-wavelength transport modes.

Finally, the seminar will survey the clever alternative paradigms designed to bypass these previous "exact" computational bottlenecks within the MD framework without explicitly computing the standard heat flux.
I will examine non-equilibrium techniques such as kinetic energy rescaling and Müller-Plathe velocity-swapping methods under periodic boundary conditions, as well as the calculation of "thermostatting work" in systems coupled to localized thermal reservoirs. This survey culminates in a detailed look at the Approach to Equilibrium Molecular Dynamics (AEMD), analyzing how the transient relaxation of an initial, periodic non-equilibrium temperature profile can be fitted to the analytical solutions of the heat diffusion equation to directly extract thermal diffusivity.
By critically evaluating the underlying merits and intrinsic limitations of each approach —balancing the rigorous quantum foundations of the BTE against the structural flexibility of Green-Kubo and classical MD— this presentation aims to provide a lucid, unifying roadmap of the current state of the art, shedding light on the subtle theoretical limits and open questions that still challenge our ability to decode heat transport in the condensed phase.