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Perspective

Ultrafast thermodynamics: Toward entropy, heat, and criticality on sub-nanosecond timescales

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ACCESSIBLE OVERVIEW Materials consist of vast numbers of ions and electrons, which can be excited and probed on ultrafast timescales. The resulting non-equilibrium dynamics of these excitations generate entropy and heat on sub-nanosecond timescales, which are the subject of study of ultrafast thermodynamics. We introduce and outline this nascent field and discuss potential directions for further development.

SUMMARY

Recent experimental advancements have enabled the control and probing of a wide variety of material excitations using ultrafast laser pulses. This development lies at the core of the emerging field of ultrafast thermodynamics, which enables the definition of heat, work, and entropy production on picosecond timescales and along individual trajectories. In this framework, non-equilibrium material excitations are treated as effective systems coupled to a stochastic environment. Here, we outline the potential of ultrafast thermodynamics using spin and lattice excitations, non-equilibrium collective phenomena, and criticality. We conclude by presenting open problems and ideas for further progress in this promising research field.

INTRODUCTION

Thermodynamics is one of the most successful and enduring frameworks in physics, with origins dating back to the early nineteenth century. Its development was closely intertwined with the Industrial Revolution, particularly through the study and optimization of heat engines. A microscopic foundation for thermodynamics was later provided by statistical physics, which relates macroscopic thermodynamic quantities—such as temperature and pressure—to statistical averages over an enormous number of microscopic degrees of freedom. Today, statistical physics plays a central role across nearly all areas of physics and beyond, proving essential for the understanding of critical phenomena both in and out of equilibrium, collective classical and quantum behaviors, and phenomena in solid-state, soft, and biological matter.

A large interacting system can often be conceptually divided into a “system” and a “bath.” The system contains the variables of interest, while the bath represents the remaining environment. If the bath relaxes quickly and couples weakly to the system, its influence can be approximated as random and dissipative, leading to a Langevin-type stochastic evolution. Brownian motion provides a classic example of this picture: the erratic motion of a mesoscopic particle arises from its interaction with the surrounding water molecules.

More recently, thermodynamics has been extended to systems characterized by significant time-dependent fluctuations and governed by stochastic dynamics. This field, known as stochastic thermodynamics,^{1–5} provides a consistent framework in which heat, work, and entropy can be defined and predicted along individual particle trajectories.^{4,6} Stochastic thermodynamics has been successfully applied to investigate the energetics of far-from-equilibrium systems—including soft, active,^{7–10} and biological¹¹ matter—as well as chemical processes¹² and quantum systems interacting with their environment.⁵

Independent of the development of stochastic thermodynamics in the past 20 years, efforts in describing the dynamics and non-equilibrium phenomena have also played an increasing role in solid-state and quantum materials.^{13–15} Here, the vast amount of ions and electrons gives rise to emergent phenomena, i.e., macroscopic properties that the individual constituents do not have. Prominent examples include various types of spin, charge, and orbital ordering; superconductivity; or topological phases of matter. Controlling these exciting phenomena with ultrashort laser pulses has emerged as a highly active research area during the past 30 years.

Here, experiments are typically based on a pump-probe scheme, or variants thereof, in which the material is excited into a non-equilibrium state by a pump pulse and time-dependently



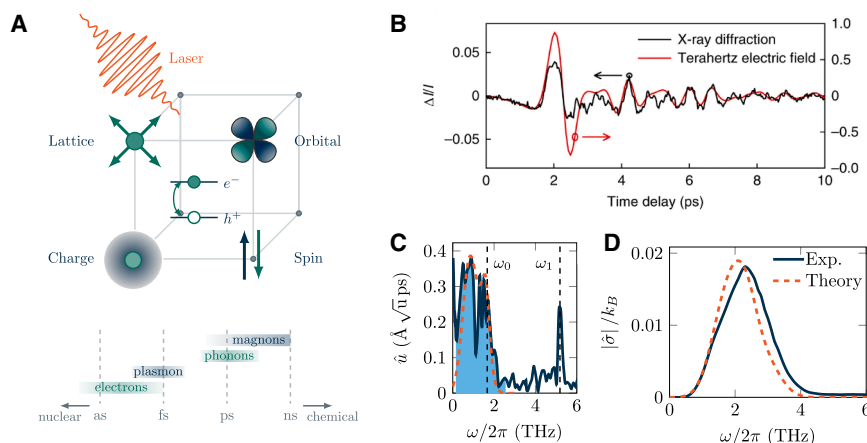


Figure 1. Ultrafast thermodynamics of material excitations

(A) Schematic illustration of various degrees of freedom in materials together with the characteristic timescale of a few examples of material excitations.

(B) Evolution of the ferroelectric soft mode in SrTiO₃, measured using time-resolved X-ray diffraction. Reprinted with permission.¹⁹ Copyright 2019, Springer Nature.

(C and D) Fourier transform of the evolution of the ferroelectric soft mode (blue) compared to a theoretical model³² (orange) (C) and Fourier transform of the corresponding entropy production rate (D). Reprinted with permission.³² Copyright 2024, the authors of the original article (licensed under CC BY 4.0).³²

probed to observe the evolution of the non-equilibrium state. The frequency, pulse width, intensity, and polarization of the pump pulse are chosen such that it selectively excites a specific mode in the material. Probe schemes can be of a different nature. A few selected examples include the magneto-optical Kerr effect used to study magnets^{16,17}; X-rays¹⁸ used for structural, electronic,²¹ and magnetic^{22,23} degrees of freedom; and transport, such as measuring anomalous Hall currents²⁴ and critical currents in superconductors.²⁵

THERMODYNAMICS ON PICOSECOND TIMESCALES

We introduce ultrafast thermodynamics, bridging the gap between stochastic thermodynamics and ultrafast materials science. The timescales of different ultrafast excitations in materials are illustrated in Figure 1A. The merger between thermodynamics and these excitations becomes most illustrative in the example of collective lattice vibrations—phonons. Infrared-active optical phonons are collective displacements of atoms inducing local dipoles, which couple to an electric field and can therefore be triggered with laser light. In contrast, Raman-active phonons are excited by non-linear optical excitation²⁶ or a non-linear phononic excitation.^{27,28} The frequency of an optical phonon lies in the THz range. Excitingly, the excitation and decay of a single phonon mode can be followed experimentally, e.g., using ultrafast X-ray diffraction^{19,29} or the THz-induced Kerr effect.^{28,30,31}

An example of experimental data¹⁹ for the so-called ferroelectric soft mode in the oxide perovskite material SrTiO₃ is shown in Figure 1B. Besides the high accuracy of measuring the evolution of the phonon mode, it becomes obvious that noise plays a crucial role in the mode dynamics. In fact, to a good approximation, the dynamics of the ferroelectric soft mode can be described by a classical Langevin equation for a single particle in a harmonic potential.³² Such harmonic approximation is valid for moderate ionic displacement.^{33,34} The thermodynamics of the ferroelectric soft mode after laser excitation should follow the rules developed in the framework of stochastic thermodynamics.^{1,32} Remarkably, the information gathered by the X-ray scattering, together with the specifics of the pump pulse, is

enough to reconstruct the entropy and heat production in the material.³² Figure 1C shows the Fourier transform of the time-resolved X-ray diffraction signal, indicating the frequency of the involved phonon mode. In Caprini et al.,³² this information was used to compute the entropy production rate in frequency space, shown in Figure 1D. We note that strong excitations of phonon modes, particularly in materials such as SrTiO₃, would require corrections for lattice anharmonicity, altering the equation of motion.^{29,35}

In the low-temperature regime, a purely classical description of the phonon dynamics becomes insufficient. Quantum nuclear effects must be taken into account to explain anomalous behaviors of soft-mode phonons, such as quantum paraelectricity.³⁶ In this quantum regime, the non-equilibrium dynamics of a THz-driven phonon and the resulting heat and entropy production have to be computed within a fully quantum framework, for example, using quantum Langevin equations for operators or appropriate quantum master equations. Consistently, the fluctuation-dissipation relation must also be replaced by its quantum version.³⁷

The stochastic evolution of the phonon mode is a direct consequence of the fluctuation-dissipation theorem. As any excitation in a material has a finite lifetime, its decay is closely connected to the emergence of noise (fluctuations) in the sample. Therefore, the stochastic evolution is observed for all types of material excitations. Besides phonons, examples could be the non-equilibrium excitation of charge (plasmons)³⁸ or spin (magnons). In fact, coupling magnetic degrees of freedom (system) with phononic degrees of freedom (bath) allows us to develop a theory of “Brownian motion” for magnets.^{39,40} Hence, entropy and heat production in laser-driven modifications of the material magnetization can also be inferred from spectroscopic information, e.g., determined using the magneto-optical Kerr effect.^{41,42}

LOSING MEMORY: SWITCHING BETWEEN NON-MARKOVIAN AND MARKOVIAN DYNAMICS

The stochastic dynamics of a material excitation, such as a phonon or magnon, is due to the interaction with the bath, effectively capturing microscopic processes in the material, including

phonon-phonon (spin-spin), electron-phonon (electron-spin), and spin-phonon interactions. When this interaction is effectively instantaneous and does not retain information about the past of the system, the resulting dynamics is referred to as Markovian. In contrast, non-Markovian dynamics is characterized by a backflow of information or energy from the bath to the system. In the following, we discuss how non-Markovianity can emerge and how it can be measured within the framework of ultrafast thermodynamics.

The Markovian approximation is valid when the intrinsic time-scale of the system (slow) is much longer than the correlation time of the bath (fast). However, if ultrafast driving accelerates the system dynamics to time scales comparable to the bath correlation time, memory effects can no longer be neglected, and non-Markovian phenomena such as information and energy backflow will emerge.

This phenomenon is best illustrated by a Gaussian laser pulse of width $\Delta\tau$, which has a bandwidth $\Delta\omega$ in the frequency domain. Since pulse width and bandwidth are inversely related ($\Delta\tau\Delta\omega = \text{const.}$), shorter pulses result in broader bandwidths. The coupling to the bath, mediated by microscopic degrees of freedom in the material, modifies the frequency-dependent structure of the system's susceptibility via bath spectral density.

While long pulses with narrow bandwidth probe only a limited frequency range, shorter pulses enable the system response to sample a broader spectrum, thereby revealing the bath spectral density. In general, however, narrow-band sources with high field amplitudes and long pulses are uncommon. Moreover, detecting a mode becomes challenging when its lifetime is shorter than the pump duration.^{43,44}

In the case of phonons, a colored dissipation kernel and noise, corresponding to a frequency-dependent bath spectral density, can be derived from a model of coupled harmonic oscillators, both classically^{45,46} and quantum mechanically within the Caldeira-Leggett model.^{37,47–49} When non-Markovian effects become dominant, a non-monotonic decay of phonon modes is expected,³⁷ as observed, for example, in LaAlO_3 .^{50–52} Recent advances in molecular dynamics simulations based on neuro-evolution potentials^{53,54} enable the simulation of laser-driven phonon dynamics. For instance, using this approach, the colored noise correlation function of SrTiO_3 has been determined.⁴⁵

Another platform for studying memory effects is ultrafast stochastic magnetization dynamics. Here, memory effects can be derived by incorporating spin-phonon interactions, referred to as “open-system” magnetization dynamics.³⁹ Excitingly, this non-Markovian behavior was recently observed in cobalt thin-film samples, using the magneto-optical Kerr effect.⁵⁵ By comparing the experimentally observed magnetization dynamics with numerical simulations incorporating an open-system kernel, it becomes evident that memory effects play an essential role. This result demonstrates that non-Markovianity is experimentally accessible. Furthermore, the experimental data show that the non-Markovian effects are only visible and relevant on ultrafast timescales because the signals vanish for longer times.

As established by the second law of thermodynamics, the total entropy production of an isolated system is non-negative. However, recent results have shown that the reduced (system-level) en-

tropy production rate may become temporarily negative on short timescales,⁵⁶ particularly when the environment is not locally equilibrated, i.e., in the presence of non-Markovian effects. This behavior does not contradict the second law but rather reflects that standard expressions for entropy production in stochastic thermodynamics neglect system-environment correlations and thus provide an incomplete description. Consequently, this quantity does not, in general, provide a faithful estimate of the total thermodynamic entropy production. Nevertheless, deviations from positivity of the entropy production rate can serve as a measure of non-Markovianity.^{37,57} Within the framework of ultrafast thermodynamics, the sign of the entropy (or heat) production rate can be inferred spectroscopically,^{32,41} enabling the exploration of the non-Markovian character of collective excitations.

ENTROPONS AND NOVEL NON-EQUILIBRIUM COLLECTIVE PHENOMENA

One of the most recent applications of stochastic thermodynamics concerns the prediction of collective excitations that arise uniquely out of equilibrium. These excitations emerge in crystalline structures in which each unit of the crystal is governed by non-equilibrium or active dynamics, for instance, generated by the continuous consumption of energy from the environment. This energy consumption is typically modeled via an active force⁵⁸ that drives the system into a non-equilibrium steady state with non-vanishing currents, not described by a Maxwell-Boltzmann distribution. In this regime, active forces generate collective excitations with a dispersion relation distinct from that of conventional thermally induced phonons.⁵⁹ These excitations constitute additional lattice vibrations, termed entropons, as they can be interpreted as waves of entropy production or, equivalently, as modes of the system's spectral entropy production.⁶⁰

So far, entropons have been theoretically predicted in soft-matter systems with solid-like order, such as high-density active colloids, where activity stems from chemical reactions,⁵⁸ and dense assemblies of active granular particles, driven out of equilibrium by internal motors. In these systems, thermal noise excites conventional phonons, while the continuous energy consumption gives rise to entropons and a finite entropy production rate in the non-equilibrium steady state. Although entropons are collective lattice vibrations, they differ fundamentally from phonons: their unique dispersion relation is determined by the microscopic properties of the active forces or the specific mechanisms driving the system out of equilibrium.⁶⁰

Besides soft-matter systems, detecting entropons at ultrafast and atomic scales remains a significant experimental and conceptual challenge. One promising platform could be topological defects in solid-state systems, where emergent active behavior in the form of self-propulsion has been proposed.⁶¹ Also, entropons could emerge whenever a crystal is traversed by a steady flux of charge, heat, particles, or energy. In such situations, the system resides in a non-equilibrium steady state characterized by broken detailed balance and continuous entropy production. In general, the identification of entropons at microscopic scales and solid-state systems, together with their experimental observation, is a promising future research direction.

METASTABILITY AND CRITICALITY

Phase transitions describe the sudden change of a macroscopic property of a system. This macroscopic property is typically described by an order parameter, such as the magnetization in a ferromagnet. In equilibrium, phase transitions are by now well understood and characterized. In addition to static triggers such as chemical doping or strain, the emergence of a specific phase in a material can also be induced dynamically using laser pumping. Looking back at the aforementioned example, the oxide perovskite material SrTiO_3 shows a tendency toward establishing a ferroelectric phase transition at low temperatures, which is believed to be suppressed by quantum fluctuations. The vicinity of the ferroelectric phase transition is connected to the softening of a Γ -point phonon—the ferroelectric soft mode. Laser driving of another higher-frequency optical phonon mode (A_{2u}) allows us to effectively strain the sample, introducing a transient and metastable phase transition to a ferroelectric phase.^{62–64}

This is just one example of a variety of laser-induced metastable phases and phase transitions observed throughout the past decades. Other examples are light-induced superconductivity,^{25,65,66} magnetism,^{67,68} charge order,⁶⁹ charge density wave,⁷⁰ and even unconventional orders such as chirality,⁷¹ where the order parameter is still under debate.⁷² While equilibrium phase transitions are well understood in the framework of the Landau-Ginzburg-Wilson theory, emerging from the minimization of a free energy functional, the same procedure is not applicable in general to non-equilibrium phase transitions and requires an extension toward a non-equilibrium theory of critical phenomena.^{73–75} These dynamical critical phenomena can be probed on ultrashort timescales, as shown, for example, by the critical slowing down in charge-ordered systems^{76,77} and charge density waves.⁷⁸ Methods based on stochastic equations of motion for the order parameter^{3,79,80} have proven promising for describing the aforementioned phenomena.

Phase transitions in active matter, complex fluids, and biological systems with irreversible behavior typically depend on microscopic dynamics. In these cases, the order parameter can be described by stochastic equations of motion, allowing one to formulate a stochastic thermodynamics at this coarse-grained level.^{3,81} This approach has already been employed to shed light on collective phenomena in active matter.⁸² Moreover, non-equilibrium phase transitions have recently been described through least-action principles based on trajectories.⁸³ This framework could provide a promising route toward modeling metastability in laser-excited materials and realizing protocols to stabilize and control phase transitions on sub-picosecond timescales.

LOOKING AHEAD: AN ULTRAFAST THERMODYNAMICS ROAD MAP

Probing the non-Markovian regime

A structured bath spectral density implies a finite correlation time of the bath but does not by itself guarantee non-Markovian dynamics. But when the relevant system timescales (e.g., intrinsic dynamics or external driving) become comparable to this bath correlation time, the evolution of the system can resolve the temporal correlations of the bath, leading to a breakdown of the

Markovian approximation. In the frequency domain, this corresponds to the fact that an ultrafast drive, which has a broad frequency distribution with significant spectral weight in the relevant frequency range, can effectively sample the frequency dependence of the system's susceptibility, which contains information about the bath spectral density. In this framework, we propose the following.

- Several experiments have explored the emergence of non-Markovianity, for example, in quantum dots^{84,85} and quantum processors.⁸⁶ In contrast, we propose to extend the study of non-Markovianity in magnetic materials. We propose to go beyond the promising example of fcc cobalt thin films⁵⁵ by employing time-resolved magneto-optical Kerr effect measurements for different magnetic materials.
- In magnetic systems, multiplicative noise naturally arises from stochastic fluctuations of the effective magnetic field generated by ordered spins on a lattice.³⁹ In contrast, additive noise corresponds to longitudinal spin fluctuations, which are often neglected, though it can be incorporated phenomenologically into the equations describing magnetization dynamics.⁸⁷ As with multiplicative noise, microscopic mechanisms underlying longitudinal noise should also be developed.
- The non-Markovian dynamics of phonons should be probed, e.g., using time-resolved X-ray diffraction or optical probes, such as the optical Kerr effect or time-resolved Raman scattering. By progressively shortening the pulse duration and thereby broadening its frequency content, different components of the phonon spectrum can be excited,³⁷ which in turn activates bath memory effects and makes them visible in subsequent phonon dynamics.
- First-principles molecular dynamics should be used to compute the effective bath spectral density for individual infrared-active or Raman-active phonon excitations. We assume that this function is not uniform but rather exhibits a structure that depends on the phonon density of states and mode-coupling strengths.

Metastability and the non-equilibrium phase transition

Stochastic thermodynamics has been extended to order parameters (fields), i.e., by using functional methods such as the Martin-Siggia-Rose-Janssen-de Dominicis formalism. Within this framework, we propose to investigate laser-excited metastable phases of matter and dynamical phase transitions through the lens of entropy production. Below, we outline two key research routes related to this topic.

- Extend the concept of trajectories to fields and derive the corresponding out-of-equilibrium least-action principles.⁸³
- To obtain a guiding example, derive the non-linear phononics description of metastability in perovskites⁶² from a more general entropy-based formulation.

Ultrafast stochastic thermodynamics to predict novel collective excitations

Stochastic thermodynamics has been employed to predict out-of-equilibrium collective excitations, known as entropons, in

soft-matter crystals where each constituent unit operates far from equilibrium. We expect that ultrafast (stochastic) thermodynamics could be used to predict and characterize collective excitations at smaller length scales. Here, we outline several promising research directions.

- Applying ultrafast thermodynamics to crystalline systems driven out of equilibrium by steady-state fluxes or currents may represent a promising route to predict entropies at the atomic scale. Such an approach may provide a quantitative framework for predicting entropies in solid-state systems and, ultimately, guiding their experimental observation.
- Typically, the notion of a particle emerges by quantizing a classical field, such as the photon as the quantization of the electromagnetic field or the phonon as the quantization of lattice vibrations. Hence, we propose the quantization of entropies, thereby providing their quantum-mechanical description.
- Explore the realization of entropies using non-equilibrium dynamics of topological defects in crystals.⁶¹
- Beyond the discovery of entropies, ultrafast thermodynamics should be employed to other systems, ranging from superconductors to strange metals, with the ambitious goal of characterizing their collective out-of-equilibrium excitations and perhaps advancing their comprehension through a suitable thermodynamics analysis.

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AUTHOR CONTRIBUTIONS

Conceptualization, R.M.G.; writing – original draft, F.T., Y.Q., L.C., and R.M.G.; writing – review & editing, F.T., Y.Q., L.C., and R.M.G.; funding acquisition, L.C. and R.M.G.

DECLARATION OF INTERESTS

The authors have no competing interests.

DECLARATION OF GENERATIVE AI AND AI-ASSISTED TECHNOLOGIES IN THE WRITING PROCESS

During the preparation of this work, the authors used LeChat and ChatGPT for proofreading and spellchecking. After using this tool or service, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

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