Colloquium: Nonthermal pathways to ultrafast control in quantum materials

Alberto de la Torre*  
Department of Physics, Brown University, Providence, Rhode Island 02912, USA

Dante M. Kennes†  
Institut für Theorie der Statistischen Physik, RWTH Aachen University and JARA-Fundamentals of Future Information Technology, 52056 Aachen, Germany  
Max Planck Institute for the Structure and Dynamics of Matter, Center for Free Electron Laser Science (CFEL), Luruper Chaussee 149, 22761 Hamburg, Germany

Martin Claassen‡  
Department of Physics and Astronomy, University of Pennsylvania, Philadelphia, PA 19104, USA

Simon Gerber§  
Laboratory for Micro and Nanotechnology, Paul Scherrer Institut, Forschungsstrasse 111, CH-5232 Villigen PSI, Switzerland

James W. McIver¶ and Michael A. Sentef∗∗  
Max Planck Institute for the Structure and Dynamics of Matter, Center for Free Electron Laser Science (CFEL), Luruper Chaussee 149, 22761 Hamburg, Germany

(Dated: March 30, 2021)

We review recent progress in utilizing ultrafast light-matter interaction to control the macroscopic properties of quantum materials. Particular emphasis is placed on photoinduced phenomena that do not result from ultrafast heating effects but rather emerge from microscopic processes that are inherently nonthermal in nature. Many of these processes can be described as transient modifications to the free-energy landscape resulting from the redistribution of quasiparticle populations, the dynamical modification of coupling strengths and the resonant driving of the crystal lattice. Other pathways result from the coherent dressing of a material’s quantum states by the light field. We discuss a selection of recently discovered effects leveraging these mechanisms, as well as the technological advances that led to their discovery. A road map for how the field can harness these nonthermal pathways to create new functionalities is presented.

CONTENTS

I. Introduction 1

II. Technical advances shaping ultrafast quantum materials science 2

A. Experimental tools 2

B. Theoretical tools 5

III. Emergent phenomena after pulsed laser excitation 7

A. Weak-excitation regime: Collective modes and disentangling degrees of freedom 7

B. Optical switching 9

C. Out-of-equilibrium critical behavior 9

D. Nonlinearities and dynamical couplings 10

1. Dynamical Hubbard $U$ 10

2. Nonlinear phononics 12

IV. Dressed states of nonequilibrium matter 12

A. Theory introduction 14

1. Dynamical localization in a driven atomic chain 14

2. Floquet primer 14

B. Dressed band structures: Floquet topological insulators 16

C. Towards Floquet many-body physics: Heating and interactions 17

D. Engineering correlated systems 19

V. Outlook 21

References 22

I. INTRODUCTION

Quantum materials host a wide range of many-body and topological phenomena that both challenge our physical understanding of solids and offer possibilities for next-generation technologies. From unconventional superconductivity to topologically protected edge modes, the remarkable physics in quantum materials emerges from complex interactions between spin, charge, lattice, and orbital degrees of freedom (Keimer and Moore, 2017)
and the geometric and topological aspects of their wavefunctions (Narang et al., 2021; Wang and Zhang, 2017). Many materials host multiple quantum phases that can be independently accessed by application of external perturbations such as electromagnetic fields, pressure, strain, or chemical doping, making these systems ideal platforms for future technological applications (Tokura et al., 2017). Moreover, the search for new ways to create and control their macroscopic properties continues to improve our fundamental understanding of the interactions between the underlying degrees of freedom, which in turn may lead to new functionalities (Alexandradinata et al., 2020; Basov et al., 2017; Giustino et al., 2021).

One promising route to controlling and understanding quantum materials is ultrafast light-matter interaction. This can induce long-lived nonequilibrium states with functionally-relevant properties that cannot be realized in thermal equilibrium. Since the pioneering work of Koshihara et al. 1990 on photoinduced phase transitions, ultrafast optical experiments along these lines have been used to investigate a variety of nonequilibrium effects in quantum materials. Breakthrough results in the past decade include the surgical decoupling of microscopic degrees of freedom, ultrafast switching into hidden phases, dynamically controlled microscopic interactions, possible light-induced superconductivity, and photon-dressed topological states. These findings have been enabled by the development of a new generation of experimental and theoretical tools to probe and understand the ultrafast processes in these systems (Sec. II).

The mechanisms behind many of the most intriguing nonequilibrium phenomena go beyond the simple melting of thermal states by laser-induced heating. Instead they rely on distinct nonthermal pathways, that is, pathways that are so fast and disruptive, or where the laser fields are so strong and strongly coupled to the material, that a quasi-thermal description with effective temperatures is insufficient or impossible.

Here we survey recent efforts in applying nonthermality as a resource to exert control over quantum materials on ultrashort timescales in a flexible and reversible manner. We highlight phenomena that emerge after pulsed laser excitation, including nonthermal switching mechanisms, out-of-equilibrium critical behavior, and the nonlinear control of microscopic couplings and phononics (Sec. III). Another class of nonthermal phenomena are those that occur during the duration of the light pulse, where the microscopic degrees of freedom are strongly coupled to the amplitude, frequency and polarization of the light field, in some cases forming dressed states that can be described by Floquet theory. While well established in atomic systems, photon-dressing effects in quantum materials can lead to fundamentally new states of matter. We review recent progress in the field towards achieving Floquet engineering in quantum materials and discuss new theoretical directions that serve as a guide for the next generation of Floquet-inspired experiments (Sec. IV).

A full-fledged review of the rapidly growing field of ultrafast materials science is beyond the scope of this Colloquium. To account for the important early works, we refer to other reviews, including Averitt and Taylor 2002, Basov et al. 2011, Orenstein 2012, Zhang and Averitt 2014 and Giannetti et al. 2016, which provide extensive reports of ultrafast spectroscopy of strongly correlated electron systems. Kirilvuk et al. 2010 wrote an in-depth review on ultrafast magneto-optical effects. Buzzi et al. 2018 reviewed ultrafast structural dynamics in solids probed by time-resolved X-ray scattering. Overviews on time-resolved inelastic X-ray scattering have been provided by Cao et al. 2019 and Mitrano and Wang 2020. Further reviews on Floquet engineering can be found in (Bukov et al., 2015; Oka and Kitamura, 2019; Rudner and Lindner, 2020).

II. TECHNICAL ADVANCES SHAPING ULTRAFAST QUANTUM MATERIALS SCIENCE

A. Experimental tools

State-of-the-art light sources can now reliably generate intense optical pulses at wavelengths spanning from the THz to the extreme ultraviolet (XUV) with a wide range of available pulse durations and repetition rates (Bartels et al., 2002; Cerullo and De Silvestri, 2003; Liu et al., 2017; Reimann, 2007; Steimleyer et al., 1999). These recent advances have enabled a more surgical approach to photoexciting (‘pumping’) quantum materials in pump-probe experiments, for example by selectively accessing particular electronic transitions, directly coupling to collective modes, or intentionally avoiding such resonances entirely. This enhanced pump tunability, combined with a fleet of updated experimental probes targeting complementary macroscopic observables (Fig. 1), has led to an improved understanding of nonequilibrium phenomena in solids and the nonthermal pathways leading to their creation. Here, we introduce some of the most widely used ultrafast techniques together with recent works that showcase their capabilities.

Time-resolved optical spectroscopy remains the most commonly used approach to access the time-dependent optical properties of quantum materials after photoexcitation. In recent years, the field has seen an evolution from the early measurements of transient absorption and reflectivity (Elsayed-Ali et al., 1987; Koshihara et al., 1990; Miyano et al., 1997; Schoenlein et al., 1987) into a multifaceted set of techniques, where the particular details of the experiment depend on the targeted subsystem and dynamics (Averitt and Taylor, 2002; Orenstein, 2012). Some of the most advantageous capabilities of these approaches are: (i) direct detection of tran-
sient changes in the electronic joint density of states via frequency-resolved measurements of the transient complex optical conductivity from the THz to the XUV range (Baldini et al., 2020; Jager et al., 2017; Sie et al., 2015; Siegrist et al., 2019), (ii) simultaneous measurements of the dynamics of different subsystems by combining multiple detection schemes (von Hoegen et al., 2018), including transient non-linear optical processes (Mahmood et al., 2021; Sala et al., 2016; Woerner et al., 2013) and polarization rotations sensitive to changes in magnetic orders (Beaurepaire et al., 1996; Kinel et al., 2020; Kirilyuk et al., 2010; Némeck et al., 2018; Schlauderer et al., 2019; Walowski and Münzenberg, 2016), (iii) the integrability with other external stimuli, for example magnetic fields and hydrostatic pressure (Cantaluppi et al., 2018; Mitrano et al., 2014; Nicoletti et al., 2018; Trigo et al., 2012), and (iv) the ability to modulate and control the optical pulse to gain real-space information (Gedik et al., 2003; Mahmood et al., 2018; Torchinsky et al., 2014). These techniques have enabled the observation of a wide range of phenomena in the time domain including quasiparticle relaxation dynamics, electron-boson coupling strengths, gap magnitudes, photoexcited order parameters and collective mode oscillations (Brorson et al., 1990; Demsar et al., 1999a, 2002; Segre et al., 2002), phase coexistence, light-induced phase transitions (Giannetti et al., 2016; Koshihara et al., 1990; Rini et al., 2007; Yusupov et al., 2010a), magnetic moment precession, (Afanasiev et al., 2021; Disa et al., 2020; Nova et al., 2017; Stupakiewicz et al., 2021), the relaxation dynamics of superconductors, and light-induced superconducting-like states (Cremin et al., 2019; Demsar et al., 1999b; Fausti et al., 2011; Hu et al., 2014; Kaindl et al., 2000; Mitrano et al., 2016; Nicoletti et al., 2014; Yu et al., 1991).

**Time-resolved scattering techniques.** Ultrafast X-ray scattering techniques take advantage of photon wavelengths of comparable magnitude to the atomic spacing to probe structural dynamics (Cavalleri et al., 2006; Fritz et al., 2007; Gerber et al., 2015, 2017; Johnson et al., 2008; Mankowsky et al., 2014; Rettig et al., 2015) or...
the evolution of spin, charge and orbital electronic orders (Beaud et al., 2014; Dean et al., 2016; Först et al., 2011; Johnson et al., 2012; Kubacka et al., 2014; Lee et al., 2012; Mitran et al., 2019) after optical excitation. The way towards sub-picosecond X-ray pulses has been paved by laser-driven (Rischel et al., 1997) and hybrid laser-accelerator-based sources (Schoenlein et al., 2000, 1996), which in addition to diffraction (Cavalleri et al., 2006; Johnson et al., 2008) have also enabled spectroscopic measurements (Cavalleri et al., 2005; Stamm et al., 2007). The advent of X-ray free-electron lasers (FEL) (Emma et al., 2010; Seddon et al., 2017) has led to another burst of quantum materials studies via femtosecond pulses of Angstrom wavelength and increased brilliancy. We refer to in-depth reviews on ultrafast X-ray scattering (Buzzi et al., 2018) and resonant inelastic X-ray scattering (Cao et al., 2019; Mitran and Wang, 2020).

The wavelength tunability of these pulsed X-ray sources is key to accessing different phenomena. For example, the ‘soft’ X-ray regime (250 – 2000 eV) includes many strongly resonant absorption edges of individual elements, e.g., 3d transition metals, which can be targeted to probe the electronic properties of materials. Although such resonances also exist in the ‘hard’ X-ray regime (> 5000 eV), its main use is to access structural properties and large portions of reciprocal space. X-ray FELs can also be used to access inelastic scattering channels in the time domain, for example to map out energy- and momentum-resolved nonequilibrium phonon dispersions (Trigo et al., 2013; Zhu et al., 2015) and dynamics of magnetic correlations (Dean et al., 2016; Mazzone et al., 2020), as well as disordering during ultrafast phase transitions (Wall et al., 2018). The coherence of FEL light has also been used to directly probe fluctuating topological order via X-ray photon correlation spectroscopy (Seaberg et al., 2017).

Ultrafast electron diffraction (UED) is a complementary scattering technique that also directly measures ultrafast structural dynamics in solids (Baum and Zewail, 2007; Siwick et al., 2003; Zewail, 2006). The larger scattering cross section of electrons, as compared to X-ray photons, makes UED particularly well-suited to studying thin samples. New MeV electron sources (Weathersby et al., 2015), THz streaking (Kealhofer et al., 2016; Zhao et al., 2018) and pulse compression schemes (Kim et al., 2020; Qi et al., 2020) can now achieve < 100 fs time resolution. Recent experiments have taken advantage of these new capabilities, e.g., to investigate lattice dynamics (Konstantinova et al., 2018; Mannebach et al., 2015; Waldecker et al., 2017) and electronic orders (Haupt et al., 2016; Le Gruyader et al., 2017; Sie et al., 2019; Vogelgesang et al., 2018; Zong et al., 2018), as well as to study electron-phonon couplings (René de Cotret et al., 2019; Harb et al., 2016; Horstmann et al., 2020; Stern et al., 2018; Waldecker et al., 2016).

Time- and angle-resolved photoemission spectroscopy (tr-ARPES) measures changes in the band structure and single-particle spectral function of solids with momentum resolution (Bovensiepen and Kirchmann, 2012; Gedik and Vishik, 2017; Haight et al., 1988; Lv et al., 2019; Nicholson et al., 2018; Petek and Ogawa, 1997; Smallwood et al., 2016; Wegkamp et al., 2014; Zhou et al., 2018). tr-ARPES has been used, for example, to study decoherence effects in the excitation process (Höfer et al., 1997; Ogawa et al., 1997; Reutzel et al., 2019), to shed light on the physics of high-temperature superconductors (Avigo et al., 2013; Parham et al., 2017; Smallwood et al., 2012; Yang et al., 2014, 2019), to track the melting and recovery of charge-density wave orders (Hellmann et al., 2010, 2012; Liggies et al., 2018; Perfetti et al., 2006; Retting et al., 2016; Rohwer et al., 2011; Schmitt et al., 2008; Zong et al., 2019b), to directly probe excitonic states (Cui et al., 2014; Madéo et al., 2020), to measure the relaxation dynamics of photocurrents (Giädde and Höfer, 2021; Reimann et al., 2018) and the coupling between electronic and lattice degrees of freedom (Gerber et al., 2017; Kemper et al., 2017; Na et al., 2019). This technique also permits the detection of transiently populated topological states (Belopolski et al., 2017; Sobota et al., 2012, 2013; Zhang et al., 2017), the observation of Floquet-Bloch states (Mahmood et al., 2016; Wang et al., 2013), and the identification of nonthermal electronic regimes (Gierz et al., 2013; Johannsen et al., 2013; Na et al., 2020). An exciting prospect is the implementation of new detection schemes extending time- and momentum-resolved microscopy to FELs (Kutnyakhov et al., 2020).

Time-resolved scanning probes. Time-resolved scanning near-field optical microscopy (tr-SNOM) tracks photoinduced changes in the optical constants of materials on the 10 nm length scale with high temporal and spectral resolution (Eisele et al., 2014; Wagner et al., 2014). tr-SNOM has been used, for example, to investigate photoinduced insulator-to-metal transitions (Dönges et al., 2016; Huber et al., 2016) and image the propagation of plasmon-polaritons in a variety of systems (Huber et al., 2017; Ni et al., 2016; Wagner et al., 2014). Ultrafast scanning tunneling microscopy (STM) is also gaining traction and probes quantum tunneling at the atomic length scale with sub-femtosecond time resolution (Cocker et al., 2013; Garg and Kern, 2020; Nunes and Freeman, 1993). Ultrafast STM has been used to track carrier and spin dynamics in photoexcited semiconductors (Terada et al., 2010; Yoshida et al., 2014), the ultrafast vibrational motion of a single molecule (Cocker et al., 2016), and image surface plasmons in gold (Garg and Kern, 2020).

Time-resolved transport. Microstructured devices incorporating laser-triggered photoconductive switches (Auston, 1975) have been used to investigate a variety of ultrafast transport phenomena, such as ballistic electron flow in carbon nanotubes (Zhong et al., 2008), helicity-dependent photocurrents in topological insulators (Kastl et al., 2015), and the transport properties of Floquet-
Bloch states in graphene (McIver et al., 2019). Ultrafast transport dynamics have also been probed in carbon nanotubes (Gabor et al., 2012), graphene (Sun et al., 2012), and various van der Waals heterostructures (Arp et al., 2019; Ma et al., 2016; Massicotte et al., 2016) by measuring the photocurrent generated in response to two time-delayed laser pulses.

B. Theoretical tools

Most statistical and computational techniques available in thermodynamic equilibrium cannot be straightforwardly generalized to systems driven far from equilibrium. This makes the theoretical description of the out-of-equilibrium microscopic dynamics of a quantum many-body system particularly challenging. Nevertheless, new experimental results have triggered complementary theoretical efforts towards modeling microscopic dynamics in photoexcited quantum materials.

Time-dependent density functional theory (TDDFT) is an extension of ground-state density functional theory, with similar merits and challenges (Marques et al., 2012; Ullrich, 2012). The full time-dependent Schrödinger equation for the many-particle wavefunction is replaced by an auxiliary set of Schrödinger equations (Kohn-Sham equations), which determine the exact time-dependent density of the system (Runge and Gross, 1984). Coulomb interactions are accounted for via exchange-correlation potentials, though their exact forms are unknown in practice. TDDFT has been successfully applied to a host of systems in weak and strong external driving fields, ranging from atoms and molecules to periodic solids (De Giovannini et al., 2013; Lian et al., 2020). The strength of TDDFT lies in the ability to describe full-fledged material-specific details. However, the faithful description of nontrivial correlation effects still poses a severe challenge for TDDFT-based methods, in particular in pump-probe settings.

Nonequilibrium Green’s functions and diagrammatic techniques provide a framework for obtaining few-body correlation functions of time-dependent problems without computing the actual many-body wave functions (Stefanucci and van Leeuwen, 2013). Green’s function techniques are based on single-particle propagators that describe the probability amplitude for particles (electrons, phonons, etc.) to travel between two space-time points while interacting with the rest of the system. These many-body interactions are described through the many-body self-energy. Notable approximations to calculate the self-energy include the so-called GW approximation for screened interactions (Aryasetiawan and Gunnarsson, 1998; Thygesen and Rubio, 2007), nonequilibrium dynamical mean-field theory (DMFT) for strongly correlated systems, including Mott insulators with mainly local self-energies (Aoki et al., 2014; Freericks et al., 2006; Tsuji et al., 2008), and the real-time functional renormalization group (FRG) approach (Kennes et al., 2012). In a nonequilibrium setup these approximation schemes are more difficult to handle than in equilibrium because time-translational invariance cannot be exploited and, in general, two time variables instead of one energy variable must be kept. Nonequilibrium Green’s functions are an ideal starting point when approximations for the self-energy can be physically motivated, e.g., perturbative weak- or strong-coupling expansions, or the local-self-energy approximation of DMFT.

Green’s function-based real-time diagrammatic quantum Monte Carlo techniques provide alternative and a priori controlled solutions via stochastic sampling of a perturbation series. While these approaches suffer from a notorious ‘dynamical sign problem’ (Werner et al., 2009), whereby the non-positivity of sampling weights results in a computational effort that increases exponentially in time, recent algorithmic advances with modest computational scaling including the ‘inchworm’ algorithm (Cohen et al., 2015) and conformal transformations (Bertrand et al., 2019) have been put forward. Real-time diagrammatic quantum Monte Carlo is the method of choice whenever the sign problem can be averted.

Tensor networks provide a faithful representation of many-body states and operators as a ‘contraction’ network of tensors that encode the local properties of the system (Schollwöck, 2011; Verstraete et al., 2008). In one dimension matrix product states are one particularly prominent example of a tensor network and provide the basis for elegant density matrix renormalization group implementations (Schollwöck, 2011). In general, the representation of generic quantum states requires contracting tensor dimensions that scale with the exponential size of the full many-body Hilbert space. However, in many physically relevant situations the notion of locality introduced by the tensor network allows one to represent states with low entanglement via tensors of much reduced dimensionality. In this sense, tensor networks might be viewed as a low-entanglement method. The entanglement of ground states generally scales favorably in low-dimensional systems due to the area law (Eisert et al., 2010; Orús, 2014). Conversely, dynamics far from equilibrium typically involve highly excited states with volume law scaling of the entanglement entropy. Therefore, in nonequilibrium studies only short time scales can be simulated. Numerous methods for time-evolution have been proposed and benchmarked, including the time-evolving block decimation, matrix product operator techniques, Krylov methods, or the time-dependent variational principle (Paeckel et al., 2019). In a nutshell, tensor networks are useful whenever entanglement entropy can be kept at bay.

Other variational techniques. Beyond asymptotically exact representations of the many-body wavefunction, simpler variational starting points serve as theoretical
tools to gain insight into specific physical systems, often based on some intuition of the relevant physics. Examples include the time-dependent Gutzwiller approximation (Schiró and Fabrizio, 2010; Seibold and Lorenzana, 2001), variational Monte Carlo (Carleo et al., 2011), Gaussian and non-Gaussian variational states (Hackl et al., 2020; Shi et al., 2018), or Gross-Pitaevskii equations (Gross, 1961; Pitaevskii, 1961). More recently, machine learning-inspired restricted Boltzmann machines have emerged, which provide a more flexible variational subspace for time evolution (Carleo and Troyer, 2016). To summarize, common to all these techniques are Ansätze for the many-body wavefunction with an economical number of free parameters, which are governed by equations of motion as determined via the time-dependent variational principle. The general usefulness of these techniques for simulations related to pump-probe experiments is not quite clear yet. However, they can provide a good starting point whenever a suitable variational manifold can be identified.

III. EMERGENT PHENOMENA AFTER PULSED LASER EXCITATION

We discuss here emergent phenomena in quantum materials occurring after photoexcitation with an ultrafast laser pulse. Comprehensive reviews of these effects can be found by Basov et al. 2011, Orenstein 2012, Zhang and Averitt 2014 and Giametti et al. 2016. In particular, we focus on processes dominated by dynamics that cannot be described by a hot electronic subsystem which thermalizes back to equilibrium through heat exchange with the cold crystalline lattice (phonons), the so-called two-temperature models (Allen, 1987; Anisimov et al., 1974) or n-temperature generalizations (Koopmans et al., 2010). These descriptions rest on the assumption that the mutual couplings between degrees of freedom are not modified by the excitation (Allen, 1987; Bauer et al., 2015; Petek and Ogawa, 1997). In quantum materials, this is not necessarily the case, and the optical excitation can result in modified or suppressed coupling constants (Ishioka et al., 2008; Murakami et al., 2015). More importantly, these simplified models neglect nonthermal effects resulting from ultrafast light-matter coupling. For example, in many correlated systems the notion of thermality, in the sense of a unique thermal density matrix and temperature, breaks down on ultrashort (femtosecond) to intermediate (sub-picosecond) time scales (Kemper et al., 2018). This departure from the thermal response makes the pulsed excitation of quantum materials a unique strategy for creating and controlling quantum phenomena.

To better summarize this rapidly evolving and complex field, we depict in Fig. 2 different scenarios of transient phenomena in terms of a simplified potential energy landscape as a function of a system coordinate, e.g., an electronic order parameter or a lattice displacement. (a) Weak excitations around a stable minimum (ground state) permit probing collective modes and their mutual couplings. (b) With a short, more intense laser pulse, one can switch between degenerate ground states (A and B), or drive the material to a nonthermal, metastable excited state C. (c) Nonequilibrium critical behavior can be induced by a short, even more intense excitation that transiently modifies the potential energy landscape itself. (d) With a strong excitation, the system can also be driven into anharmonic regimes with deviations from parabolic behavior, which allows to probe nonlinear effects and change effective couplings in the material.
A. Weak-excitation regime: Collective modes and disentangling degrees of freedom

We consider a situation where the system under investigation is only weakly driven, i.e., it remains in a linear regime [Fig. 2(a)]. The recovery towards equilibrium is sensitive to collective modes and reflects the characteristic timescales of the different degrees of freedom. Prominent examples of coherent modes in ordered states that have been probed using time-domain techniques are amplitude modes of charge-density wave materials (Demsar et al., 1999a, 2002; Lee et al., 2012; Mihailovic, 2019; Schmitt et al., 2008; Zong et al., 2019a), potential excitonic insulators (Werdehausen et al., 2018), and the Higgs amplitude (Chu et al., 2020; Matsunaga et al., 2013, 2014) and Leggett modes (Giorgianni et al., 2019), as well as Josephson plasmons (Laplace and Cavalleri, 2016) in superconductors.

In the weak-excitation regime, charge, lattice, spin or orbital degrees of freedom of correlated electron systems can be disentangled in the time domain by appropriate choice of the experimental probe [Fig. 3(a)]. Under particular circumstances these techniques can be brought together in a unified fashion, as demonstrated by Gerber et al. 2017 where time-resolved ARPES and X-ray scattering were used to study the significance of a cooperative interplay among electron-electron and electron-phonon interactions in the iron-based superconductor parent compound FeSe. Figure 3(b) depicts how both the lattice displacement and the electronic band structure lock into a coherent $1_g$ phonon mode that is driven by an ultrafast IR laser pulse. In turn, this THz frequency locking of the two degrees of freedom allows for a purely experimental, highly precise quantification of fundamental physical properties. For example, for FeSe it was used to quantify the orbitally-resolved electron-phonon deformation potential, which reveals the critical importance of electron correlations for the material’s properties.

This example shows that such multi-messenger approaches not only allow inferring nonequilibrium characteristics, but also to extract equilibrium properties via the time domain (Gerber et al., 2017; Mitrano et al., 2019). A caveat concerns the difficulty in bringing together more than one ultrafast experiment in a unified fashion. Ideally, a multi-messenger experiment is conducted in one single setup, e.g. as used for simultaneous detection of electronic and structural orders (Porer et al., 2014) or multiple structural order parameters (Kogar et al., 2020; Morrison et al., 2014; Zhou et al., 2021). This ensures that the pumping conditions, such as pump fluence, pump and probe beam profile, time resolution, penetration depth (effectively absorbed energy densities) and, notably, also the definition of time zero (the instant when the pump pulse excites the sample) are guaranteed to be consistent. Eventually, such unified experimental setups will not only allow comparing the amplitudes signals of the disentangled degrees of freedom, but also their phase relation and decay rates (dephasing times), paving the way towards coherent quantum control of (dis-)entangled degrees of freedom.

B. Optical switching

Going one step further away from equilibrium, a natural question to ask is whether nonthermal pathways can be found that bring the system to parts of the free-energy landscape that are not accessible in thermal equilibrium. The key idea is similar to basic concepts of chemical reac-
Recent work suggests that the underlying pathways in which catalysts overcome an activation energy barrier in order to trigger reactions.

We start with a pedagogical example of the complex phases emerging from competing ground states in quantum materials, as sketched in Fig. 2(b). We consider a long-range ordered symmetry-broken ground state with two or more possible lowest-energy states $A$ and $B$. They can represent, for example, two opposite spin configurations of a ferromagnet, the left- and right-handed chiral groundstates of a topological superconductor or the respective ground states of two competing orders. Without lack of generality, we can also assume the existence of other local minima $C$ in their vicinity. Once the system has thermalized into a minimum of the free-energy landscape, for example $A$, reaching $B$ or $C$ has an associated energy cost and might not be possible in thermal equilibrium. However, excitation with an ultrashort laser pulse can overcome this energy barrier. In experiments, the observation of optical switching requires the absorption of a sufficiently short pulse – to prevent thermalization before the switching process has happened – and the subsequent relaxation of the photoexcited system into a metastable state via dissipation of the surplus energy into a heat bath (phonons, substrate). The equilibrium state is, in general, recovered by either a remarkably slow relaxation process or upon increase of temperature. A recent comprehensive theoretical description in terms of a Ginzburg-Landau theory with relaxation dynamics can be found in Sun and Millis 2020.

The possibility to access long-lived states with desired properties has motivated an increasing number of ultrafast experiments in materials with free-energy landscapes that fall within the simplified description of Fig. 2(b). In the following we discuss examples of optical switching experiments in which the transfer of energy into the electronic degrees of freedom results in some nonthermal distribution needed to access the metastable state. For example, in the correlated insulator $\text{VO}_2$, the excitation of electronic degrees of freedom, via either a pump resonant with an absorption transition (Baum et al., 2007; Becker et al., 1994; Cavalleri et al., 2004, 2001; Morrison et al., 2014; Otto et al., 2019; Wall et al., 2018; Wegkamp et al., 2014) or THz fields in nanofabricated samples (Liu et al., 2012), causes a structural transition into a metallic phase, not accessible at low temperatures. In manganites, the coupling of orbital, spin and lattice degrees of freedom results in an antiferromagnetic insulating ground state (Tokura and Nagaosa, 2000) from which a metallic phase can be accessed by either laser (Beaud et al., 2014; Ichikawa et al., 2011; Li et al., 2013; Takubo et al., 2005; Teitelbaum et al., 2019; Zhang et al., 2016) or electric field (Asamitsu et al., 1997; Miyano et al., 1997) pulses. Moreover, theoretical predictions suggest that the creation of charge photoexcitation in the presence of the strong spin-orbital exchange interactions in the manganites can lead to a partial melting of these competing intertwined orders that guides the material into a new metastable phase (Li et al., 2018).

In the layered dichalcogenide $1T'$-$\text{TaS}_2$ (Fazekas and Tosatti, 1979), the ultrafast transfer of energy into the electronic degrees of freedom, by either laser (< 35 fs) (Danz et al., 2021; Densmar et al., 2004; Haupt et al., 2016; Hellmann et al., 2012; Ligges et al., 2018; Perfetti et al., 2006; Stojchevska et al., 2014; Zong et al., 2018) or electrical (∼ 30 ps) pulses (Vaskivskyi et al., 2015) results in a long-lived metallic state, as highlighted in Fig 4 (a). Recent work suggests that the underlying mechanism of transition into the metallic state and subsequent metastable amorphous-like electronic order (Gerasimenko et al., 2019; Vodeb et al., 2019) is the formation of topological defects due to light-induced disor-
der (Haupt et al., 2016; Laulhé et al., 2017). In prototypical Peierls systems, a highly non-thermal electronic density of states can lead to an ultrafast transition into a long-lived high-symmetry phase (Huber et al., 2014; Naseska et al., 2020; Teitelbaum et al., 2018) [Fig. 4(b)]. In the 1/8-doped cuprates, superconductivity is suppressed due to competition with charge (stripe) order. Excitation with an infrared pulse leads to a long-lived state with superconducting-like properties at higher temperatures than at equilibrium (Cremin et al., 2019; Fausti et al., 2011; Nicoletti et al., 2014). In the charge density wave (CDW) compounds, RTe$_3$ (R = lanthanide), ultrafast switching into a competing CDW state inaccessible in thermal equilibrium has been realized after photoexcitation (Koger et al., 2020; Zhou et al., 2021). Ultrafast laser pulses have also been used to switch the topological state of a material, as demonstrated in THz-driven WTe$_2$ (Sie et al., 2019).

New approaches towards flexible switching strategies with fast and reversible recoveries, for example unimpeded by the healing of topological defects (Mihailevic, 2019), are being theoretically and experimentally explored. In the search for novel switching mechanisms for future technological application, the field of multiferroic materials – in which electric fields can for instance control magnetism – has served as inspiration (Spaldin, 2020). Ultrafast switching into a metastable ferroelectric phase has been observed after coherently driving the quantum paraelectric phase of SrTiO$_3$ with strong THz fields (Li et al., 2019; Nova et al., 2019). This approach has also been used to demonstrate the ultrafast reversibility of the ferroelectric polarization in both directions – key for future applications (Mankowsky et al., 2017; Qi et al., 2009). In multiferroic heterostructures mid-IR excitation has been used to engineer a coherent three-dimensional structure with polar, strain and charge-ordering with nanometer periodicity (Stoica et al., 2019).

Finally, we discuss the key ideas behind symmetry-guided switching (Chou et al., 2017; Dasari and Eckstein, 2019; Dehghani et al., 2020; Dehghani and Mitra, 2017; Kennes et al., 2019a; Ono and Ishihara, 2019; Sentef et al., 2017; Tsuji and Aoki, 2015), in particular, the example of a chiral superconductor (Claassen et al., 2019). Here the phases A and B in Fig. 2(b) are the right- and left-handed chiral groundstates. It was shown that a combination of linearly polarized followed by circularly polarized laser pulses can nudge the system from the right-handed to the left-handed chiral groundstate. Importantly, the system can immediately be switched back equally fast by applying another sequence of pulses and simply changing the chirality of the circularly polarized laser pulse. The important ingredient besides laser polarization is the duration of the two pulses: the circular pulse has to be applied while the system has not yet recovered from the linear pulse and remains in a superposition of the two chiral states. Recently, it was shown how the same mechanism but with spatially localized laser spots could be used to write, move, or erase chiral domains in real space (Yu et al., 2021).

C. Out-of-equilibrium critical behavior

Ultrafast photoexcitation can induce an instantaneous change of the free energy landscape by suppressing the relevant order parameter [Fig. 2(c)]. This phenomenology challenges our understanding of symmetry-breaking phase transitions. In equilibrium, the macroscopic time-dependent Ginzburg-Landau (GL) theory assumes an order parameter which varies ‘sufficiently’ slowly in time. However, when the order parameter is suppressed on system-intrinsic timescales, it is unclear whether equilibrium concepts (universality classes, scaling) remain applicable, and how the order parameter fluctuations evolve after light excitation. Here we discuss out-of-equilibrium symmetry-breaking phase transition both as a pathway towards accessing nonthermal effects but also as a platform to study fundamental questions about macroscopic properties associated with order parameter dynamics. The seminal works by Hohenberg and Halperin 1977, Polkovnikov et al. 2011 and Täuber 2017 offer comprehensive reviews of this topic.

An example of universal critical behavior in out-of-equilibrium phase transitions is the slowing down of the recovery dynamics. Similarly to what occurs in thermal equilibrium, in the vicinity of a second-order transition, spatial fluctuations of the order parameter diverge and it takes an exponentially long time to reinstate the ordered phase. This has been seen, for example, in the electronic recovery of charge order in perovskite manganites (Beaud et al., 2014) and La$_{1/3}$Sr$_{2/3}$FeO$_3$ (Zhu et al., 2018), as well as in the spin and orbital dynamics in YVO$_3$ and GdVO$_3$ (Yusupov et al., 2010b), and LaVO$_3$ (Lovinger et al., 2020). Divergent slow dynamics also characterize the ultrafast suppression of the nematic electronic order in FeSe (Shimojima et al., 2019) and in iron pnictides (Patz et al., 2014). This seemingly universal behavior in out-of-equilibrium second-order phase transitions has been discussed in the time-dependent GL framework (Dolgirev et al., 2020), by stochastic Langevin equations describing the relevant subsystems (Sieberer et al., 2016), and by non-equilibrium Green’s functions (Schüler et al., 2018).

Additionally, ultrafast phase transitions open the possibility to study the effects of nonadiabatic suppression. Near the phase transition the ordered and disordered phases are quasi-degenerate in energy. This allows the fluctuations of the order parameter to be arbitrarily slow. Within the Kibble-Zurek mechanism (Kibble, 1976; Zurek, 1996), when the order parameter suppression happens on timescales faster than that of the fluctuations,
the coherence between order-parameter domains is lost and the critical behavior is replaced by recovery dynamics dominated by the healing of topological defects. This behavior is universal (Bray, 1994) and has been observed in the long-time recovery dynamics after suppression of the striped phase in cuprates (Mitrano et al., 2019) and of the charge order in LaTe3 (Zong et al., 2019b) [Fig. 5(a)], TbTe3 (Mertelj et al., 2013; Yusupov et al., 2010a), and 1T-TiT2 (Vogelgesang et al., 2018). A different nonequilibrium scenario has been shown to emerge when two strongly coupled order parameters are quenched by a laser: in the striped nickelate La1.75Sr0.25NiO4 an absence of topological defects was reported in conjunction with slow phase fluctuation recovery (Chuang et al., 2013; Lee et al., 2012) and explained by a time-dependent GL theory (Kung et al., 2013).

We finally discuss predictions of nonthermal behavior in strongly correlated systems when driven through a phase transition (Chiocchetta et al., 2017). For example, quenching of the electron-electron repulsion U in the antiferromagnetic phase of the Hubbard model results in a suppression of the magnetization. Depending on the size of the quench it was shown that a nonzero order parameter and coherent amplitude mode oscillations survive even for quenched values of U at which the system would be in the disordered phase in thermal equilibrium (Balzer et al., 2015; Sandri and Fabrizio, 2013; Tsuji et al., 2013; Werner and Murakami, 2020; Werner et al., 2012). This gives rise to a prototypical out-of-equilibrium phase diagram shown in Fig. 5(b), where the suppression of the amplitude mode is associated with a secondary nonthermal critical point at $U_\text{f} = U_\text{nth}^\text{crit}$. It is worth noting that such a transition is not expected within the GL picture, where the amplitude mode oscillations vanish as the curvature of the free-energy potential is suppressed and, consequently, the restoring force disappears (Hohenberg and Halperin, 1977).

D. Nonlinearities and dynamical couplings

We now discuss cases where nonlinear regimes are reached through intense excitation and showcase two examples: (i) dynamical effective interactions due to modified electronic screening, in which the nonlinearity stems from electron-electron interactions, and (ii) strongly driven crystal lattices, in which the nonlinearity is due phonon-phonon interactions or other phonon-related nonlinearities. In both cases the effective Hamiltonian is transiently modified.

1. Dynamical Hubbard U

The dynamical regime after an ultrafast photoexcitation opens a pathway for changing the effective interactions in correlated materials. The paradigmatic example for such an effective interaction is the Hubbard U. Simply speaking, U parametrizes the screened local part of the Coulomb integrals of strongly localized (typically d or f) orbitals. Examples for applications of the Hubbard model are transition-metal oxides (Imada et al., 1998). There, localized transition-metal ions are embedded between more delocalized ligand orbitals, which often are energetically separated from the d orbitals. In such cases...
a downfolding procedure can be applied (Miyake et al., 2009), in which the low-energy effective model contains a Hubbard $U$ that depends on the screening from electrons in delocalized environmental orbitals. The key idea of dynamical Hubbard $U$ is that the electronic excitations created after interaction with an ultrafast laser pulse can modify the screening environment, effectively changing $U$ within femtosecond time scales [Fig. 6(a)].

To explain the underlying mechanism of dynamical Hubbard $U$ we discuss here the transient reduction of $U$ upon photoexcitation in the correlated antiferromagnetic charge-transfer insulator NiO predicted within time-dependent density functional theory plus $U$ (TDDFT+U) (Tancogne-Dejean et al., 2018). The underlying physics can be understood as follows: The electronic structure of NiO is such that the localized correlated subspace of mainly $d$-orbital character and the rest of the system of mainly $p$-orbital character can be considered independently. Photoexcitation of electrons from the localized subspace into delocalized states results in additional screening. This naturally leads to a decrease of the effective $U$. It is instructive to think of the excitation as an induced time-dependent polarization of the localized electrons, affecting the dielectric properties of the delocalized electrons, which is expected for strong driving fields even for frequencies away from optical transitions. Similar results were obtained by Gillmeister et al., 2020 and Golež et al., 2019 using different methods.

Experimental evidence for a dynamically modified Hubbard $U$ by photoexcitation of electron-hole pairs was reported in the transition-metal dichalcogenide MoTe$_2$ (Beaulieu et al., 2021). In this material, the uncorrelated band structure has two electron-like bands crossing the Fermi level $E_F$ at the edge of the Brillouin zone. However, this is not observed in photoemission. Instead, as explained by DFT+$U$, the Hubbard $U$ pushes the respective states above $E_F$. This effect can be partially undone on ultrafast time scales, as demonstrated by tr-ARPES. At a time delay of $\sim 200$ fs after photoexcitation, an ultrafast Lifshitz transition was observed, at which the Fermi surface topology changed because the electron-like bands were pushed below $E_F$ triggered by dynamical reduction of $U$.

A different route towards dynamical modulation of Hubbard $U$ is the coherent driving of the crystal lattice. Dynamical $U$ through phonon driving was first demonstrated in a prototypical one-dimensional Mott-insulating charge transfer salt (Kaiser et al., 2014a; Singla et al., 2015) and more recently reported as a microscopic mechanism for possible photomolecular superconductivity far above equilibrium $T_C$ in two-dimensional molecular crystals (Buzzi et al., 2020; Tindall et al., 2020). The phononic route towards dynamical interactions is particularly suited for molecular crystals, where the effective local sites of the Hubbard model are comprised of entire molecules or parts thereof, which can be vibrationally excited and the on-site effective interaction thus changed [Fig. 6(b)] through the deformation of the intra-molecular wavefunction during the vibration. The key advantage of resonant phonon excitation in the mid-IR frequency range in comparison to photodoping by optical pulses lies in the frequency selectivity of the excitation pathway. This brings us right to the discussion of vibrational control of emergent phenomena in quantum materials in the following subsection.

FIG. 6 Dynamically screened Hubbard $U$ in laser-driven quantum materials. (a) Screening is typically enhanced when a material is excited by an optical laser pulse, which creates electronic excitations, resulting in a dynamically reduced time-dependent $U$ (Tancogne-Dejean et al., 2018; Golež et al., 2019). In charge-transfer insulators this results in a narrowing of both the Mott-Hubbard gap between the upper and lower Hubbard bands (UHB and LHB, respectively), as well as the charge-transfer gap $\Delta$ (Tancogne-Dejean et al., 2020). (b) For a midinfrared laser excitation, resonant with an infrared-active phonon mode, the dynamical $U$ parametrically depends on the phonon mode coordinate $Q_{IR}$ via a nonlinear electron-phonon interaction ($Q_{IR}^2U$) (Buzzi et al., 2020; Singla et al., 2015). In this case, whether $U$ is increased or decreased on average depends on the sign of this nonlinearity.
an IR and a Raman (R) mode, with a coupling strength $g_{\text{cubic}}$ that is determined by the anharmonic potential. Coherent driving of the IR mode to large amplitudes [Fig. 7(a)], using strong midinfrared laser pulses, couples to the Raman modes such that a unidirectional displacive force occurs that is proportional to the square of the IR-mode displacement [Fig. 7(b)]. The consequences of the respective transient crystal structures [Fig. 7(c)] on the electronic subsystem have been shown to bear potential for changing the phases of quantum materials on ultrafast time scales in various ways (Mankowsky et al., 2016).

Among others, coherent control of the crystal lattice by driving to anharmonic regimes has been suggested as a mechanism to induce ultrafast phase transitions, such as a lattice-controlled metal-insulator transition (Rini et al., 2007), and lead to states of matter without equilibrium counterparts, including possible light-induced superconductivity (Fausti et al., 2011; Hu et al., 2014; Kaiser et al., 2014b; Mankowsky et al., 2014). In turn, these groundbreaking experiments have stimulated considerable theoretical activity on nonequilibrium superconductivity (Babadi et al., 2017; Coulthard et al., 2017; Dasari and Eckstein, 2018; Denny et al., 2015; Kennes et al., 2017; Kim et al., 2016; Knap et al., 2016; Komnik and Thorwart, 2016; Mazza and Georges, 2017; Murakami et al., 2017; Nava et al., 2018; Okamoto et al., 2016; Raines et al., 2015; Sentef, 2017; Sentef et al., 2016, 2017; Wang et al., 2018a), as well as a lively debate on how we should understand and interpret optical superconducting-like signatures on transient time scales (Demsar, 2020; Zhang et al., 2020).

Furthermore, coherent optical driving of the lattice has been suggested as a path towards switching applications (see Sec. III.B) as well as a means of mapping of interatomic forces and potentials (von Hoegen et al., 2018; Kozina et al., 2019). Another developing subfield is phono-magnetism (Afanasiev et al., 2021; Disa et al., 2020; Giorgianni et al., 2021; Juraschek et al., 2020; Nova et al., 2017; Stupakiewicz et al., 2021), in which magnetic properties of materials are modified by coherent phonon driving.

To summarize, phononic control of quantum materials is a rapidly growing research field, conceptually located at the boundary between ultrafast dynamics after short laser excitation and dressed states of nonequilibrium matter, which will be discussed in the following.

### IV. DRESSED STATES OF NONEQUILIBRIUM MATTER

Another class of nonequilibrium phenomena encompasses the effects that occur in solids during optical illumination. Examples include high-harmonic generation (Ghimire et al., 2011; Schubert et al., 2014; von Hoegen et al., 2018), sub-cycle electron and spin dynamics (Kampfrath et al., 2011; Lucchini et al., 2016; Reimann et al., 2021).
FIG. 8 Versatile functionalities enabled by Floquet engineering. (a) Periodic driving using optical fields can steer the macroscopic properties of quantum materials by dressing their microscopic degrees of freedom, such as electronic hopping parameters. (b) Examples include proposals and experimental realizations of new electronic band structures (Mahmood et al., 2016; Wang et al., 2013), the manipulation of band topologies (Kitagawa et al., 2011; Lindner et al., 2011; McIver et al., 2019; Oka and Aoki, 2009), and the observation of excitonic Stark shifts caused by the creation of ultrastrong synthetic magnetic fields (Kim et al., 2014; Sie et al., 2015). Additional proposals include the optical control of magnetic correlations (Claassen et al., 2017; Kennes et al., 2018; Mentink et al., 2015), experimentally observed in quantum simulation settings (Görg et al., 2018), the creation of fractionized quasiparticles obeying nonabelian statistics (Lee et al., 2018), and forms of photon-dressed superconductivity (Kennes et al., 2019a; Knap et al., 2016; Murakami et al., 2017; Tindall et al., 2020).

et al., 2018; Schultze et al., 2013), light-field driven currents (Higuchi et al., 2017; Schiffrin et al., 2013) and the parametric amplification of plasma waves (Rajasekaran et al., 2016). Comprehensive reviews of many of these topics can be found by Ghimire and Reis 2018 and Kampfrath et al., 2013. Here, we focus on observables described by the coherent dressing of a material’s quantum states by the rapidly oscillating light-field. The manner and degree to which the states are dressed are determined by the amplitude, frequency and polarization of the electromagnetic drive. Such coherent light-matter interaction thereby provides an external control knob for transiently tuning the electronic and magnetic interactions in solids, which can be used to engineer quantum states with no equilibrium counterpart.

The consequences of photon dressing have long been investigated in the context of atomic systems and have led to applications in atomic clocks, precision spectroscopy, and quantum information processing (Scully and Zubairy, 1997). At the root of many of these phenomena is the AC Stark effect, a process whereby the oscillating electromagnetic field effectively shifts or splits electronic energy levels via the electric dipole interaction (Autler and Townes, 1955). This fundamental dressing effect has also been observed in solid state systems using the strong AC fields in ultrafast laser pulses (Bonitz, 2016; Fröhlich et al., 1985; Haug and Jauho, 2008; Mücke et al., 2001; Reutzel et al., 2020; Rossi and Kuhn, 2002; Schäfer and Wegener, 2002; Sie et al., 2015).

Quantum materials can be dressed by light in more complex ways than isolated atomic systems by exploiting the additional degrees of freedom that interact with the light field. While this enhanced complexity is an opportunity for realizing novel material responses, it comes at the cost of increased dissipation, which leads to decoherence due to electron-electron, electron-phonon and other scattering mechanisms. Far stronger optical fields are thereby required to overcome these sources of deco-
herence in driven solid-state electronic systems (Aeschlimann et al., 2021; Nuske et al., 2020; Sato et al., 2019), which have historically made dressed states difficult to experimentally access.

Advances in the generation of strong-field laser pulses now make it possible to routinely access regimes where dressing effects dominate the response of optically driven quantum materials, which are being investigated with the aid of a new generation of ultrafast experimental probes (McIver et al., 2019; Wang et al., 2013). Theoretical breakthroughs utilizing Floquet theory (Lindner et al., 2011; Oka and Kitamura, 2019; Rudner and Lindner, 2020) are simultaneously offering unprecedented guidance to experimentalists. This combined effort has led to remarkable discoveries and predictions (Fig. 8). We review some of these developments in the sections that follow.

A. Theory introduction

To introduce the concept of dressed states in solids, we begin with the intuitive example of a periodically driven atomic chain in the high-frequency limit. We then generalize to the case of arbitrary frequency, for which Floquet theory provides a flexible and powerful framework to describe dressing effects.

1. Dynamical localization in a driven atomic chain

Consider a one-dimensional tight-binding model of noninteracting spinless fermions in second quantization

\[ H = -\sum_n t_0 c_n^\dagger c_{n+1} + \text{H.c.} = \sum_k \varepsilon(k)c_k^\dagger c_k , \]

where \( t_0 \) is the hopping amplitude and \( c_n^\dagger \) (annihilates) a fermion on site \( n \). In momentum space, one obtains a dispersion relation \( \varepsilon(k) = -2t_0 \cos(ka) \), where \( a \) is the lattice constant.

Next consider a coupling to a time-periodic longitudinal electric field \( E(t) = E_0 \sin(\omega t) \) with a field strength \( E_0 \) and frequency \( \omega \), which can be included by the Peierls substitution \( t_0 \rightarrow t_h(t) = t_0 e^{i e A(t)/\hbar} \) where \( A(t) = E_0/\omega \cos(\omega t) \) is the vector potential, such that \( E(t) = -\partial_t A(t) \). When the driving frequency is larger than the intrinsic energy and time scales in the system, the high-frequency oscillations can be averaged out. Time averaging \( t_h(t) \), denoted by \( \langle \ldots \rangle \), yields \( t_{\text{eff}} = \langle t_h(t) \rangle = t_0 J_0(F) \), where \( J_0 \) is the zeroth-order Bessel function and we introduced the dimensionless Floquet parameter

\[ F = \frac{aeE_0}{\hbar \omega} \]

to parameterize the strength of the electromagnetic drive, with \( e \) the elementary charge. Since \( |J_0(x)| < 1 \), the resulting effective hopping amplitude \( t_{\text{eff}} \) is always smaller than the equilibrium value \( t_0 \), meaning that on average (i.e., in the effective or dressed picture) the electrons in the driven chain are more localized than in the undriven chain. This real-space description of dynamical localization is depicted in Fig. 9(a).

Equivalently, in momentum space, the effect of dynamical localization is captured by the minimal coupling \( k \rightarrow k - eA(t)/\hbar \) in the band dispersion. The effect of the electric field is to periodically shake the dispersion to the left and right [bottom row of Fig. 9(a)]. When considering the minimum of the equilibrium dispersion relation at \( k = 0 \), one finds that this shaking on average shifts the bottom of the band dispersion to higher energy in the effective picture. Analogously, the dispersion maxima at the zone boundaries are lowered in energy upon averaging. The net effect in the effective/dressed picture is a reduction in bandwidth when compared to the equilibrium dispersion relation. This example demonstrates how periodic driving provides a tuning knob for materials properties via the renormalization of electronic hopping (Bucksbaum et al., 1990; Claassen et al., 2017; Dunlap and Kenkre, 1986; Kennes et al., 2019a, 2018; Mentink et al., 2015).

2. Floquet primer

Floquet theory provides a powerful basis to treat a periodically driven quantum system beyond high- or low-frequency expansions. Such a system is governed by the time-dependent Schrödinger equation:

\[ i\hbar \frac{d}{dt} |\Psi(t)\rangle = H(t) |\Psi(t)\rangle , \]

with a time-periodic Hamiltonian \( H(t + T) = H(t) \) with period \( T \). Floquet 1883 showed that the solution to Eq. (3), \( |\Psi(t)\rangle \), can be written as:

\[ |\Psi(t)\rangle = e^{-i\epsilon(t)/\hbar} |\Phi(t)\rangle , \]

where the \( T \)-periodic wavefunction \( |\Phi(t)\rangle = |\Phi(t + T)\rangle \) and the quasi-energies \( \epsilon_l \) are determined by the algebraic equation

\[ (\epsilon_l + n\hbar\omega) |\Phi_n\rangle = \sum_{n'} H_{n'n'} |\Phi_{n'}\rangle \]

with frequency \( \omega = 2\pi/T \) and Fourier decompositions \( H_{nn'} = \frac{1}{T} \int_0^T dt e^{i\omega t} H(t) \) and \( |\Phi_n\rangle = \frac{1}{\sqrt{T}} \int_0^T dt e^{i\omega t} |\Phi(t)\rangle \). This maps a periodic differential equation to an algebraic (quasi-equilibrium) problem. The time-dependent problem is transformed into an effective static Hamiltonian by considering \( \Phi_{\vec{k}} = \langle \ldots | \Phi_{\vec{k}^{-1}} | \Phi_{\vec{k}}^0 \rangle | \Phi_{\vec{k}}^1 \rangle \ldots \rangle^T \) and

\[ \Phi_{\vec{k}} = e^{i\Phi_{\vec{k}}} , \]
FIG. 9 Dynamical localization in an atomic chain and Floquet sidebands. (a) Illustration of dynamical localization in an atomic chain depicted in real and momentum space. The two different columns represent the periodically driven, real time picture as well as the effective, time-averaged picture. When the equilibrium band dispersion (black) is driven left and right by the light field (red), the total bandwidth is reduced in the effective (i.e. dressed) picture (blue), corresponding to a decrease in the effective hopping amplitude in real space. This band narrowing is described by the Bessel function $t_{\text{eff}} = \langle t_0(t) \rangle < t_0$ shown in the inset to the upper right panel. (b) Visual representation of the formation of Floquet replicas according to Eq. (6). At high frequency $\omega \to \infty$, the effective Hamiltonian is given by the time average only, $H^0$ [panel (a)]. At finite frequency Floquet sidebands at energy distance $\omega$ become accessible via the higher moments $H^1, H^2, \ldots$ of the Fourier series of the time dependent Hamiltonian $H(t)$ (color coding corresponds to Eq. (6)).

The Hamiltonian $H$ consists of an infinite number of copies of the time-averaged Hamiltonian $H^0$ energetically shifted by integer multiples of the driving frequency $\omega$. The higher harmonics $H^1, H^2, \ldots$ couple the nearest, next-nearest, … neighboring copies in this effective language. In the context of solids, where the undriven system is described by Bloch bands, these copies, or Floquet replicas, lead to Floquet-Bloch bands repeating periodically in momentum (Bloch) as well as energy (Floquet) space [Fig. 9(b)]. This insight resulted in the notion of side bands emerging under periodic driving early on, which can be used to alter the physical properties of a system dramatically (Kennes et al., 2019b; Oka and Aoki, 2009; Rudner and Lindner, 2020) (Fig. 10). Although Floquet theory was introduced more than 100 years ago, its implementation within new theoretical approaches (Sec. II.B) is a subject of intense research. Green’s functions, other diagrams, or effective single-particle pictures, such as used in DMFT, FRG, or DFT, can rather easily exploit these ideas (Aoki et al., 2014; De Giovannini et al., 2013; Eissing et al., 2016; Tsuji et al., 2008). For tensor networks (Kennes et al., 2018; Sahoo et al., 2019) or other variational techniques this is substantially complicated by the fact that Floquet theory requires the determination of the full spectrum.

Floquet engineering is challenging in quantum materials owing to the large optical fields ($10^7 - 10^8$ V/m) and long wavelengths needed to obtain a sizeable Floquet parameter (Eq. (2)). This necessitates the use of strong-field ultrafast laser pulses and limits current experiments to the low-frequency regime. While Floquet theory inherently assumes continuous wave driving, experimental results obtained using laser pulses with multiple optical cycles have nevertheless been well-described using this framework (Kim et al., 2014; Mahmood et al., 2016; McIver et al., 2019; Sie et al., 2015; Wang et al., 2013). We review some of these experimental results in the following section.
FIG. 10 Topological Floquet engineering in Dirac systems. (a) A coherent interaction with circularly polarized light was predicted to induce a photon-dressed topological band structure in graphene characterized by protected chiral edge states (Kitagawa et al., 2011; Oka and Aoki, 2009). (b) When the Floquet sidebands form they hybridize at their crossing points, opening band gaps ($\Delta_1$, $\Delta_2$). A gap also opens at the Dirac point ($\Delta_0$) due to a two-photon absorption/emission process that breaks time-reversal symmetry. The resulting Floquet-Bloch bands are fully gapped and possess a nonzero total Berry curvature and Chern number. (c) Real space schematic of the two-photon absorption/emission hopping process that induces a gap at the Dirac point in graphene. (d) Time-resolved ARPES measurements on Bi$_2$Se$_3$ observing the formation of Floquet-Bloch bands from the Dirac surface states. Reprinted from Mahmood et al. 2016. (e) Nonequilibrium anomalous Hall conductance of graphene driven by circularly polarized light as a function of the equilibrium Fermi level ($E_F$). The red and blue shading correspond to when $E_F$ was gated into the regions where the gaps $\Delta_0$ and $\Delta_1$ were predicted to appear based on Floquet theory. Reprinted from McIver et al. 2019.

B. Dressed band structures: Floquet topological insulators

Optically dressed quantum materials garnered considerable interest after they were proposed capable of hosting topologically nontrivial states that could be manipulated by adjusting the light field (Kitagawa et al., 2011; Lindner et al., 2011; Oka and Aoki, 2009). Such systems are often referred to as “Floquet topological insulators,” and in some cases they are characterized by topological invariants that have no equilibrium counterpart (Kitagawa et al., 2010; Rudner et al., 2013).

The first proposal along these lines was put forward by Oka and Aoki 2009. Using Floquet theory it was shown that an anomalous Hall effect could be induced in graphene by dressing its quantum states with circularly polarized light [Fig. 10(a)] (Oka and Kitamura, 2019). The physics can be summarized as follows: Floquet-Bloch bands form and hybridize at their crossing points, opening band gaps [$\Delta_1$, $\Delta_2$ in Fig. 10(b)]. A second-order process also causes a gap to open at the Dirac point ($\Delta_0$) because the interaction breaks time-reversal symmetry. The resulting fully-gapped Floquet-Bloch bands have a nonzero Berry curvature distribution that causes charges to undergo an anomalous Hall effect. By reversing the helicity of the light, the polarity of the induced Berry curvature distribution, and hence the direction of the net anomalous Hall current, can be controlled.

In the high-frequency limit, where the Floquet sidebands do not overlap and thus there are no gaps due to
band crossings, the Hamiltonian for the driven system is

\[ H_{\text{eff}} \approx H_0 + \frac{[H_{-1}, H_1]}{\hbar \omega} \quad (7) \]

where the first term is the equilibrium Dirac Hamiltonian and the second term is responsible for the gap opening at the Dirac point. Specifically for a two-dimensional Dirac cone with energy-momentum relation \( \epsilon(\vec{k}) = \hbar v_F |\vec{k}| \), with Fermi velocity \( v_F \), one obtains \( \Delta_0 = \hbar^{-1} e^2 v_F^2 E_0^2 / \omega^3 = \hbar^{-1} e^2 v_F^2 A_0^2 / \omega \), which scales quadratically with the peak electric field \( E_0 \) and is enhanced for small laser frequencies.

Kitagawa et al. 2011 interpreted Eq. (7) as the energy difference (\( \Delta_0 \)) between two processes, where an electron in the \( n = 0 \) Dirac cone first absorbs (emits) a circularly polarized photon to enter the \( n = 1 \) (\( n = -1 \)) sector, then emits (absorbs) a photon of the same helicity to return to the \( n = 0 \) sector [Fig. 10(b)]. In real space, this corresponds to a double hopping in the unit cell that has the net effect of introducing chiral intra-sublattice tunneling elements to the effective Hamiltonian [Fig. 10(c)]. The effective Hamiltonian is identical to that proposed by Haldane for a topological Chern insulator in graphene (Haldane, 1988), which was previously believed to be inaccessible via any realistic experimental method. When only the lower Chern band is populated, a topological phase is expected to develop hosting a quantized anomalous Hall effect carried by chiral edge states, which would be photon-dressed in this case.

Floquet topological insulators have been studied in the high-frequency limit using a variety of quantum simulation platforms, most notably in photonic waveguides (Rechtsman et al., 2013), where the propagating edge mode was directly observed, and in driven optical lattices of ultracold fermions (Jotzu et al., 2014). Later theoretical works confirmed that topological states could also be created in the low-frequency limit [e.g. Fig. 10(b)], with the difference being that the multitude of Floquet-Bloch bands that form due to the additional gap openings can have Chern numbers \( |C| > 1 \), and thus have multiple edge modes (Claassen et al., 2016; Dehghani et al., 2015; Mikami et al., 2016; Sentef et al., 2015; Usaj et al., 2014). An additional topological invariant dubbed the winding number can also arise in the low-frequency limit (Kitagawa et al., 2010; Rudner et al., 2013), which is defined over time rather than momentum space and is thus only realizable in periodically driven systems. Observations of such ‘anomalous Floquet topological insulators’ have also been reported in the quantum simulation community (Kitagawa et al., 2012; Maczewsky et al., 2017).

In a landmark paper, Wang et al. 2013 reported the first spectroscopic evidence of topological Floquet-Bloch bands in a solid within the Dirac surface states of the topological insulator Bi\(_2\)Se\(_3\) using tr-ARPES [Fig. 10(d)]. The data reveal multiple dressed sidebands and the opening of band gaps, similar to those illustrated in [Fig. 10(b)].

McIver et al. 2019 reported the first electrical transport results from topological Floquet-Bloch bands. Using an ultrafast transport device (bottom center panel of Fig. 1), an anomalous Hall effect was detected in graphene illuminated by a midinfrared pulse of circularly polarized light. Remarkably, at high laser pulse fluences they observed that the anomalous Hall conductance saturated on the order of two conductance quanta—the same value predicted in the high-frequency limit (Kitagawa et al., 2011). When the graphene Fermi level was tuned away from charge neutrality using an electrostatic gate [Fig. 10(e)], features were observed in the conductance spectrum closely aligned with the opening of the band gaps \( \Delta_0 \) and \( \Delta_1 \), as predicted by Floquet theory for their laser pulse parameters. Subsequent numerical studies confirmed that the observed photocurrents originated from the formation of topological Floquet-Bloch bands (Nuske et al., 2020; Sato et al., 2019). These results are an encouraging sign that Floquet-engineered topological edge states are a distinct possibility in optically driven Dirac materials.

C. Towards Floquet many-body physics: Heating and interactions

The experiments by Wang et al. 2013 and McIver et al. 2019 demonstrate that Floquet engineering in quantum materials is realistic despite the presence of strong dissipation, which leads to decoherence. However, the tr-ARPES data also reveals significant pump-induced carrier excitation, leading to nonequilibrium dressed electron distribution in the Floquet-Bloch bands. Here, distributional and dissipative effects play an important role already even in the absence of electronic interactions (Dehghani et al., 2014; Iadecola et al., 2015; Seetharam et al., 2015), and contribute for instance to substantial deviations from quantization of the Hall conductivity and edge state transport in Floquet band topological insulators (Farrell and Peregrine-Boanita, 2016; Kundu and Seradjeh, 2013). The roles of heating and thermalization hence can be expected to take on an outsized role as “Floquet engineering” is extended towards the control of interacting many-body states of matter. Therefore, materials and driving protocols need to be chosen carefully in order to minimize heating and carrier redistribution such that a nonequilibrium state proximal to the ground state of the effective Hamiltonian is realized.

Theoretically there have been multiple proposals to address the challenge of heating. In general, closed periodically driven interacting systems continuously absorb energy and heat to a featureless infinite-temperature steady state by virtue of the eigenstate thermalization hypothesis for nonintegrable systems (D’Alessio and Rigol, 2014). However, tailored drive protocols can be chosen to sup-
FIG. 11 Routes to avoid runaway heating. (a) Generic interacting systems will heat up upon applying a continuous drive, detrimental to coherent control. However, by tuning the frequency of the drive to a spectral gap in the system, this heating can in principle be pushed to (exponentially) large times. Control of prethermal states can then be achieved on intermediate to long time scales till finally thermalization will set in. (b) Examples of condensed matter systems for which such a strategy could be applied. The upper two and bottom left subpanels show Mott and charge transfer insulators (LHB and UHB denote the lower and upper Hubbard band, respectively). Another interesting example are quantum Hall insulators (lower right subpanel). Between the different Landau-Levels (LL) energy gaps proportional to the externally applied magnetic field emerge. Frequency detuning to the sharp resonances between LLs could allow for prethermal state Floquet control of the system.

press heating at short times and realize a potentially long-lived “prethermal” regime. Here, a central ingredient is a separation of energy scales, and thus a separation of time scales, in the driven system, which entail an ergodic obstruction to fast energy absorption. After an initial ramp-on period governed by fast switch-on processes, the intermediate time (period-averaged) dynamics can saturate to a prethermal plateau which is governed by an effective Hamiltonian that captures a controlled Floquet modification, leading to observable consequences for instance in correlation functions or spectroscopic probes of the system. This regime is bounded by a time scale that signifies the ultimate onset of heating towards a featureless high-temperature state [Fig. 11], which can in principle nevertheless retain certain correlations when constrained by exact nonabelian symmetries (Tindall et al., 2019).

Most of our current understanding regards the limit of high-frequency driving (Claassen et al., 2017; Gulden et al., 2020; Haldar et al., 2018; Machado et al., 2020, 2019; Mentink et al., 2015; Peronaci et al., 2018; Vajna et al., 2018; Weidinger and Knap, 2017), where the pump frequency exceeds local energy scales. In this limit rigorous results for slow heating were recently established (Abanin et al., 2017a,b, 2015; Ho et al., 2018; Kuwahara et al., 2016; Mori et al., 2018, 2016). Notably, these works provide only mathematical bounds for the onset of heating to featureless states at infinite temperature, which can be exceeded in specific settings. Many-body localized (MBL) systems constitute an important exception (D’Alessio and Polkovnikov, 2013; Lazariades et al., 2014, 2015; Ponte et al., 2015): Here, heating can be averted via a lack of ergodicity, stabilizing a tantalizing array of bona fide nonequilibrium phases, such as time-crystalline orders or Floquet topological phases (Decker et al., 2020; Rovny et al., 2018; Zeng and Sheng, 2017).

In practice, a high-frequency driving limit is absent in most real materials, since higher-lying bands and collective excitations provide a multitude of possibilities for resonant absorption at higher laser frequencies. Similarly, while MBL is realizable in cold atomic systems (Singh et al., 2019), it is typically destabilized in solids due to energy dissipation to the lattice or other degrees of freedom. Nonetheless, sufficiently slow energy absorption, for instance due to off-resonant driving or other ergodic obstructions to heating, can realize “prethermal” dynamical regimes at short times (Fig. 11), which can harbor intriguing emergent phenomena. Therefore, a central problem is to identify materials and driving regimes for which comparatively long-lived prethermal regimes persist in a realistic setting. Notably, these arguments imply that Floquet control of quantum phases in solids hence entails a two-fold challenge: (i) engineering a prethermal Hamiltonian that captures the desired dynamics on sufficiently long time scales, and (ii) engineering the electronic distribution with respect to this
prethermal Hamiltonian on the same time scales.

A complementary route to Floquet engineering aims to exploit dissipation in open systems to stabilize a driven steady state at long times, for which the energy influx absorbed from the pump is compensated by energy dissipation into the environment (Esin et al., 2018; Seetharam et al., 2019). In a solid-state setting, the crystal lattice can act as a thermostat for the electronic system due to the large time scale separation between electronic and lattice dynamics, with early works suggesting routes towards the controlled dissipative population of single-particle Floquet states (Delghani et al., 2014; Iadecola et al., 2015; Seetharam et al., 2015). The phase diagrams of infinite-time steady states of clean systems have been established to exhibit rich phase transitions (Klöckner et al., 2020; Mendoza-Arenas et al., 2017; Mitra et al., 2006; Peronaci et al., 2018; Walldorf et al., 2019), which are, however, expected to be first order at any finite driving frequency (Mathey and Diehl, 2019).

D. Engineering correlated systems

A tantalizing prospect concerns utilizing tailored light pulses to engineer novel phases of matter in correlated electron systems. Here, the use of light to modulate interactions or selectively break symmetries to tune the interplay of competing phases promises a rich playground to stabilize new phases of matter but remains a largely unexplored and methodologically challenging regime. We discuss in the following different examples.

Magnetic Mott insulators have recently emerged as a promising class of candidate materials for Floquet engineering in correlated systems (Bukov et al., 2016; Claassen et al., 2017; Kennes et al., 2018; Mentink et al., 2015; Walldorf et al., 2019). In a Mott insulator, strong local Coulomb repulsion – typically in localized transition-metal d orbitals – freezes local charge degrees of freedom at commensurate filling to open an insulating gap and form local magnetic moments. If the pump frequency is sufficiently red-detuned from the charge gap, energy cannot be absorbed resonantly as photons couple to charge [Fig. 12(a) and (b)]. Remarkably, while the charge sector remains inert on short time scales, a prethermal regime can emerge which is characterized by transiently modified effective magnetic interactions between the local moments (Claassen et al., 2017; Mentink et al., 2015).

A minimal model to describe these effects is the driven half-filled single-band Hubbard model

\[ H(t) = -t_h \sum_{\langle ij \rangle \sigma} e^{iA(t) \tau_{ij}/\hbar} \hat{c}_{i \sigma}^\dagger \hat{c}_{j \sigma} + U \sum_i \hat{n}_{i \uparrow} \hat{n}_{i \downarrow}, \]  

(8)

where \( t_h \) and \( U \) denote hopping of electrons between neighboring sites and local Coulomb repulsion, respectively, and the optical pump enters via minimal coupling to a gauge field \( \mathbf{A}(t) \).

In equilibrium, the half-filled Hubbard model at low energies and large \( U \) canonically maps onto effective nearest-neighbor Heisenberg interactions \( J \approx \frac{4t_h^2}{U} \) which arise from virtual exchange. Suppose that the pump frequency \( \omega \) is sufficiently red-detuned from the Mott gap \( \Delta \sim U - 4t_h \). If the charge sector remains inert out of equilibrium, then a straightforward perturbative calculation, which simultaneously integrates out charge degrees of freedom and photons, leads to a transient photoinduced renormalization of spin-exchange interactions, proportional to \( J \approx \sum_m \frac{4t_h^2 E_m}{U - m\hbar \omega} \) (Chaudhary et al., 2019; Itin and Katsnelson, 2015; Mentink et al., 2015).

A physical picture readily emerges by noting that the electron participating in the exchange process can now lower the Coulomb repulsion energy \( U \) of the virtual intermediate state by absorbing a photon from the pump field, thereby enhancing the effective spin exchange. A schematic depiction of this process is shown in Fig. 12(c). Conversely, for very strong pump fields, higher-order multi-photon processes can dominate and even permit flipping the sign of Heisenberg exchange if \( m \)-photon processes with \( m \hbar \omega > U \) dominate the exchange process (Mentink et al., 2015). For a typical Mott antiferromagnet with \( U \approx 2eV, t_h \approx 0.1eV, a \approx 3A \) (corresponding to \( J \approx 20meV \) in equilibrium), and pumping at 700nm, one finds an enhancement of the exchange coupling in the range \( \Delta J \sim 0.2meV \) to 20meV for a peak field strength of 0.05V A\(^{-1}\) to 0.5V A\(^{-1}\) (Batignani et al., 2015).

Frustrated quantum magnets, encompassing those materials in which competing magnetic interactions render minimizing the free-energy landscape nontrivial, are another interesting class to study. In these compounds, transient modifications of the magnetic interactions are expected to have a sizeable effect. While the role of overall renormalization of Heisenberg interactions discussed above is reflected solely in shifts of the excitation spectrum without affecting the groundstate, strongly frustrated systems typically host rich phase diagrams of competing magnetic orders such that a small photoinduced transient change of competing subdominant interactions could ‘nudge’ the system into a proximal phase (Kennes et al., 2019a). Moreover, such materials are highly sensitive to the breaking of symmetries, which grants a powerful nonequilibrium handle to steer the nonequilibrium phase on prethermal time scales, which can be kept sufficiently long via detuning from the Mott gap.

From these considerations, one avenue of utilizing Floquet engineering to transiently stabilize a novel correlated state of matter builds upon the observation that pumping a frustrated Mott insulator with circularly polarized light permits the selective breaking of time-reversal symmetry (TRS) and inversion without breaking the SU(2) spin-rotational symmetry of the magnetic
moments (Claassen et al., 2017; Kitamura et al., 2017). Notably, while TRS can be readily broken in equilibrium via external magnetic fields, the dominant contribution to low-energy spin dynamics comes from a Zeeman splitting that breaks SU(2), quickly magnetizing the system. Instead, the role of the pump is to generate a transient photoinduced scalar spin chirality $\mathbf{S} \cdot (\mathbf{S} \times \mathbf{S}_k)$, whose form can be inferred from symmetry considerations alone, and microscopically arises from fourth-order virtual processes involving elementary triangles of the underlying lattice [Fig. 12(d)]. It was shown that in Kagome antiferromagnets, such as herbertsmithite, a weak optical pump tuned to lie between one- and two-photon resonances of the parent Mott insulator can induce a transition to a proximal chiral spin liquid phase (Claassen et al., 2017), suggesting a novel nonequilibrium pathway to one of the earliest and most elusive candidates of a gapped quantum spin liquid with topological order.

Strong spin-orbit coupling multi-orbital compounds constitute a natural generalization of such a nonequilibrium separation of charge and spin degrees of freedom (Hejazi et al., 2019; Liu et al., 2018), which could allow for Floquet engineering while mitigating heating. In the orthorhombic titanates, the partially-filled $t_{2g}$ manifold has been proposed as a prime target for optical manipulation of effective Kugel-Khomskii spin-orbital interactions. In charge transfer insulators such as the cuprates, photoinduced rotational symmetry breaking was proposed to engineer magnetic interactions and ultimately destabilize the Mott antiferromagnetic phase in favor of a $d$-wave superconductor (Kennes et al., 2019a). Conversely, theoretical predictions of spectroscopic signatures suggest that the controlled manipulation of magnetic exchange interactions could be observed in time-resolved Raman or resonant inelastic X-ray scattering (Wang et al., 2018b), for instance via tracking the softening of bimagnon excitations (Wang et al., 2018c).

Fractional quantum Hall systems are another intriguing direction to engineer prethermal correlated phases using external driving. Here, while a vast series of Abelian filling fractions have been experimentally realized (Stormer et al., 1999), more elusive fractional quantum Hall states (Moore and Read, 1991; Read and Rezayi, 1999) with non-Abelian quasiparticles are well-known to prefer fine-tuned or multi-body interactions for stabilization (Greiter et al., 1991), while simultaneously incurring tremendous interest for potential applications in topological quantum computing and beyond (Nayak et al., 1995).
et al., 2008). In principle, Landau levels in ultraclean electron gases provide a natural separation of energy scales for Floquet engineering. If the pump frequency is sufficiently detuned from a cyclotron resonance, the regularity in Landau level spacing guarantees the absence of multi-photon resonances at low photon numbers. If heating can be suppressed, such a setting has been predicted to give rise to photoinduced three-body interactions that arise from virtual inter-Landau-level scattering (Lee et al., 2018), suggesting a nonequilibrium routine to stabilize a Moore-Read state. Conversely, resonant excitation between Landau levels has been proposed to engineer effective multi-layer fractional quantum Hall models (Ghazaryan et al., 2017).

V. OUTLOOK

In this Colloquium, we discussed a set of results that provide important insights into ultrafast phenomena in photoexcited quantum materials. These advances rely on an unprecedented range of optical excitations and a growing set of tools to access, probe and simulate nonthermal pathways towards control and functionality on ultrafast timescales. In particular, photoinduced phase transitions to thermally inaccessible states by short optical excitation or photon dressing constitute promising routes towards achieving nonequilibrium functionality.

Recent studies of photoinduced phase transitions have provided a more detailed understanding of the relevant microscopic degrees of freedom and their nonthermal behavior, and have revealed the interplay between these degrees of freedom in the time domain. The realization of new applications based on optical switching into metastable states is already being explored. For instance, the photoinduced structuring of $1T$-TaS$_2$ emerges as a new avenue for memory devices in low-temperature circuitry (Mraz et al., 2021). Similarly, advances in optical switching of superconductors (Budden et al., 2021) could be integrated in future microelectronics. In order to expand the range of applications, it will be necessary to identify further materials whose nonthermal energy landscape allows for efficient switching mechanisms into metastable states.

The creation and control of properties through light-matter coupling via Floquet engineering is another promising approach. In order to realize robust functionality and devices, it is necessary to understand the role of dissipation and find strategies to mitigate heating and decoherence. As such, a firm understanding of the coupling to environmental degrees of freedom is necessary. Moreover, suitable candidate materials need to be identified which host appropriate electronic structures and interactions to minimize energy absorption at selected frequencies while still permitting a controlled modification of the salient electronic features.

However, in order to reach the final goal of new functionality based on quantum dynamics far from equilibrium, it will be of increasing importance to (A) design materials tailored towards enabling precise nonequilibrium control by integrating materials synthesis and nonequilibrium experimentation synergistically and to (B) establish further bridges between the existing ultrafast material science community and adjacent research fields. For example, the young field of polaritonic chemistry is bridging nonequilibrium quantum chemistry, quantum optics, and nanoalplasmonics (Ebbesen, 2016; Feist et al., 2018; Flick et al., 2018; Ruggenthaler et al., 2018). Inspired in part by these efforts, cavity material science is an emergent research field at the boundary between quantum optics and nonequilibrium material science (Hübener et al., 2020; Juraschek et al., 2019). The key idea is the replacement of a strong laser by a few-photon state in a strongly light-matter-coupled cavity that enables control of the material properties. One possible route towards cavity control could employ the quantum nature of photons for effective Floquet engineering without the need for intense laser fields (Sentef et al., 2020).

Controlled synthesis of van der Waals heterostructures has recently emerged as a versatile tool to establish electronic and structural properties with unprecedented control and variety. In this context, twistronics – Moiré potential engineering by twisting adjacent layers – might play a crucial role. The twist angle provides a critical handle on designing electronic and structural properties which can be geared with great flexibility (Kennes et al., 2021). Crucially, the relevant kinetic energy scales of the Moiré superlattice can be orders of magnitude smaller than in a conventional material, suggesting that external nonequilibrium perturbations can have an outsized effect in determining electronic phases. Combining ultrafast light-matter interaction with twist control of such band structures hence suggests a particularly promising route towards nonequilibrium functionalization, with first explorations already under way (Kennes et al., 2021; Rodriguez-Vega et al., 2020; Topp et al., 2019). These efforts should of course be complemented by more traditional avenues of material science, such as controlled crystal growth, but now tailored specifically to questions relevant to nonequilibrium control. For example a systematic exploration of engineerable free-energy landscapes, particularly suitable for control via photoexcitation, in complex oxides is highly desirable.

These approaches expose the need for new paradigms of materials synthesis and predictions that address the requirements for ultrafast control of nonthermal states of matter. Theory needs to turn from toy models to realistic materials, to guide not only the process of directed materials synthesis, but also to provide principles for how nonthermal pathways of ultrafast control can be obtained. To achieve this leap, inspiration can
be drawn from other fields, where such an integrated approach has already been established. Namely, an increased feedback between experimental characterization, theoretical understanding, and materials synthesis is a successful strategy that is employed in many branches of quantum materials research, for instance in the field of oxides (Coll et al., 2019), or in the study of quantum materials with angle-resolved photoemission spectroscopy (Sobota et al., 2020). This interdisciplinary approach requires additional effort, as a common language and concepts need to be developed for thinking about the underlying physics. Parts of this program have been laid out in this Colloquium. This is but one step along the long path of unifying themes and languages with the goal of fostering the relatively young field of ultrafast quantum materials science.

Acknowledgments. We are grateful to H. Aoki, R. Averitt, M. Bonitz, A. Cavalleri, M. Dean, P. Hofmann, D. Juraschek, P. Kirchmann, A. Kogar, W.-S. Lee, D. Mazzone, M. Mitrano, P. Narang, D. Nicoletti, B. Normand, H. Petek, J. Ravnik, G. Refael, R. Tuovinen and A. Zong for stimulating discussions and critical feedback. DMK, JWM and MAS acknowledge support from the Max Planck-New York City Center for Non-Equilibrium Quantum Phenomena. DMK acknowledges the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) for support through RTG 1995 and under Germany’s Excellence Strategy - Cluster of Excellence Matter and Light for Quantum Computing (ML4Q) EXC 2004/1 - 390534769. MC acknowledges support from a startup grant from the University of Pennsylvania, and from the Flatiron Institute, a division of the Simons Foundation. JWM acknowledges support from the Cluster of Excellence ‘CUI: Advanced Imaging of Matter’ of the Deutsche Forschungsgemeinschaft (DFG, DFG, German Research Foundation) - SFB-925 – project 170620586. MAS acknowledges financial support through the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation) via the Emmy Noether program (SE 2558/2).

All authors contributed equally to this work.

REFERENCES
Balzer, K., F. A., Wolf, I. P., McCulloch, P., Werner, and


Demsar, J., B. Podobnik, V. V. Kabanov, Th. Wolf, and D. Mihailovic (1999b), “Superconducting gap $\Delta$, the pseudogap $\Delta_p$, and pair fluctuations above $T_c$ in overdoped Y$_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ from femtosecond time-domain spectroscopy,” Phys. Rev. Lett. 82, 4918.


metric amplification of a superconducting plasma wave," Nat. Phys. 12, 1012.


44. 19.


