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Towards Properties on Demand in Quantum Materials

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Abstract*: Over the last ten years, there has been a significant surge in the study of quantum materials. Notable findings include the prospect and identification of new Landau-symmetry-broken phases in correlated electron systems, topological phases in systems with strong spin-orbit coupling, and materials platforms based on two-dimensional van der Waals crystals that are extremely manipulable. One of the main objectives of contemporary condensed-matter physics is to find ways to experimentally realize quantum phases of matter and control their properties. This field holds promise for the development of new electronic/photonic devices with functionalities that are currently unattainable and probably unimaginable. In this Review, we discuss new approaches that are being developed to deliberately alter the parameters of microscopic interactions, which may be used to change materials into the desired quantum state. Recent advances in the use of strong fields, impulsive electromagnetic stimulation, nanostructuring, and interface engineering to modify the characteristics of electronic interactions will be highlighted in particular. When combined, these methods provide a possible road map for the age of demand-driven quantum events.*

Keywords: Quantum Technology, Material Engineering.

I. INTRODUCTION

The field of quantum materials is expanding. This phrase encompasses a wide range of substances and phenomena for which the demonstrable genuine effects of quantum mechanics exist. One reason why quantum materials are at the forefront of modern physics is that they provide an extraordinary environment for revealing the many functions of dimensionality, symmetry, topology, and strong correlations in macroscopic observables. Here, our goal is to investigate the methods and strategies for generating novel states of matter in quantum materials and adjusting their phases by external inputs. To fully use quantum advantages in new photonic, electrical, and energy technologies, practical management of these features is necessary. This is a problem with enormous social implications1. Transition metal oxides, Fe- and Cu-based high-Tc superconductors, vander Waals semiconductors, topological insulators, Weyl semimetals, and graphene are the types of quantum materials on which we shall primarily concentrate.

Quantum materials have very high sensitivity to outside influences. Interactions related to orbital, lattice, spin, and charge degrees of freedom are often comparable to electronic kinetic energy in these systems. External inputs may easily affect the fairly delicate balance between coexisting and competing ground states, resulting in a variety of quantum phases and transitions between them. Moreover, certain categories of stimulated quantum states are direct outcomes of coherent interaction between matter and light.4-6. As an alternative, direct manipulation of the electronic wavefunction and attendant Berry phase, which give rise to the anomalous electron velocity in a solid, may pre-program the features of quantum materials. Because of these supplementary control channels, research may now go beyond simple observation. Instead, by guiding a quantum material towards a desired ground, metastable, or transitory state, it is now possible to achieve "properties on demand" in a predictable manner.

Why properties on demand?

The properties-on-demand strategy's major drivers are briefly discussed in Fig. 1. First, new state of matter discoveries always revolutionize physics. Floquet-Bloch states form when intense laser pulses and surface states in Bi_2Se_3 crystals hybridize (Fig. 1a and Box 1, panel c).10,11. Floquet-Bloch states are experimentally identified by duplicates of the original electronic levels and energy gaps at avoided momentum space crossings. The Floquet technique controls Landau symmetry breaking and topological phase transitions that require a change in a global topological invariant like the Chern number.

Second, the properties-on-demand technique may solve long-standing riddles. Experiments at high magnetic fields may help solve some of the most pressing concerns in high-Tc superconductivity, such as the lack of a complete theoretical explanation14. Unresolved issues include the origin of the quantum critical point (QCP), a zero-temperature phase transition using pressure or doping, the electronic state at T > Tc that leads to superconductivity, and the ground state at T→0 without superconductivity. (Fig. 1b). QCPs are found in organic materials, Fe-based pnictides, heavy fermion systems, and (antiferro)magnetic metals15. The QCP in cuprates is difficult to access due to a superconducting dome around the putative T = 0 transition, but in a high-H field, the dome may be "removed." Depending on magnetic field intensity, these research identified quantum oscillations (QO) with various transport characteristics. A strong Fermi surface and well-defined quasiparticles are exhibited by QOs, which were originally disputed in high-Tc superconductors but are now nearly standard.

Exploiting quantum materials' anomalously strong reactions to weak stimuli is another active area. Making a Mott transistor illustrates this. Solid-state implementations of biologically inspired circuits require memory effects rooted in electronic/structural phase separation and/or electronic correlations, which are closely related to quantum materials. Memory effects may also enable energy-efficient computing. Berry phase that controls topo-logical conducting channel tuning provides another control approach. Quantum materials with hexagonal lattices, such as graphene and transition metal dichalcogenides, need the same physics for optical valley degree of freedom management. Topological protection against backscattering may also modify the chirality of electronic and photonic phenomena in quantum materials, such as chiral currents and propagating chiral hybrid light–matter modes: polaritons. Berry phase effects also account for ‘shift currents’ an optically induced charge separation caused by electronic wavefunction asymmetry which opens up new avenues for designing high-performance optical-frequency conversion and photovoltaic materials. Recent investigations of transition metal monopnictide-based Weyl semimetals with the required band topology have shown large second-order non-linear optical responses. The companion Review by Tokura et al. discusses another potential device idea.

Ways and means of quantum control

We briefly discuss the many quantum phases and state-of-the-art approaches (Fig. 2) that may be used to fine-tune a quantum material's free energy environment.

Static exogenous disturbances. These are the most controlled property adjustments because they preserve thermal balance. Hydrostatic pressurization using diamond anvil cells is used to improve orbital wavefunction overlap between crystal sites. Kinetic energy to potential energy grows. Pressure may adjust a material's Mott insulator-to-metal or superconductor34 phase barrier, causing an orders-of-magnitude resistivity change.

Nonlinear phononics. In theory, a material's symmetry may be altered by light-induced lattice displacements, which can also drastically affect the material's free-energy landscape and create new, low-energy quantum states. More specifically, net structural distortions have been imparted and/or relieved with the use of mid-infrared pulsed stimulation of infrared-active phonons. This new technique, known as nonlinear phononics, capitalizes on the coherent coupling of lattices and light. Owing to anharmonic coupling between infrared and Raman-active modes, the oscillating field modifies the lattice. "Revealing hidden phases and new states of matter" discusses spectacular photoinduced events that are facilitated by nonlinear phononics.

Metastable states. Generally speaking, impulsively produced electronic phases have lifetimes that are comparable to the material's energy relaxation time or electromagnetic pulse width. However, impulsive stimulation may trap the system in new phases at auxiliary free-energy minima in quantum materials with corrugated free-energy landscapes, which are isolated from the real ground states by a sizable kinetic barrier. On experimental timeframes, these metastable phases last forever, although they can be controlled by outside factors like temperature and magnetic field.

Floquet parameter. The electromagnetic field's amplitude or frequency may be utilized to regulate hopping, however this may conflict with Keldysh tunneling effects when $\gamma < 1$. In tightly coupled systems, magnetic exchange interactions, proportional to hopping squared, become extremely adjustable60. Floquet engineering has modulated

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ultracold atomic systems by vibrating the photonic lattice below. We have yet to employ flux engineering in solids, where external electromagnetic radiation modulates. The necessity for ultrashort laser pulses to achieve large driving amplitudes makes floquet phases fleeting and hard to distinguish. Additional questions remain concerning heating impacts mitigation and Floquet band steady-state occupancy management. Recent advances in ultrafast experimental methodologies, theoretical simulations of time-domain experiments, and population and thermal management driving algorithmsare daringly advancing this field. Time-resolved angle-resolved photoemis-sion spectroscopy (tr-ARPES) enabled the imaging of a Floquet band structure in Bi2Se3), an exciting initial step in this enormous project.

Valley control and Berry phase modulation. The Berry curvature of Bloch states in momentum space modifies the semiclassical dynamics of electrons in external fields, resulting in an anomalous velocity that gives rise to phenomena like the anomalous Hall effect. Materials having two valleys in their electronic structure, like graphene and monolayer TMDs, produce Berry curvatures of the same magnitude but opposite sign. One way to manipulate the net Berry curvature and consequently anomalous velocity related phenomena is to use off-resonant electromagnetically produced a.c. Stark shifts or valley-selective resonant optical pumping. Using flux engineering of band structures is another method put forth to manipulate Berry phase effects. In this method, dynamical breaking of time-reversal symmetry in systems such as graphene, or reorganization of orbital textures in systems such as spinorbit coupled semiconductors are used to induce topologically trivial to non-trivial transitions in systems.

Creating macroscopic quantum coherence

Perhaps the most spectacular manifestation of a macroscopic quantum process in nature is the condensation of bosons. The archetypal of quantum condensation in solids is superconductivity resulting from Cooper pairing with long-range phase coherence. Superconductivity research has produced unmatched insights and advancements in quantum matter. Condensation is a concept that applies to bosonic excitations other than superconductivity, such as magnons, excitons, and exciton-polaritons. With the ultimate objective of "condensation on demand," all of these condensates are susceptible to the complete range of parameter modification in Fig. 2, enabling the controlled investigation of new phases. Improving the highest temperature at which a certain condensate stays stable is another objective. We review briefly some of the significant developments in quantum condensates.

Magnon condensates. Building on the extensive relationship between quantum spins and Bose–Einstein condensates (BEC)88,89, these condensates represent a well-established issue. Magnon condensates may be produced via non-equilibrium excitation or those that are thermodynamically accessible. Examples of the former are achieved by tuning the magnon density to attain the dilute limit BEC with the application of a magnetic field. Among the first findings are the examples of antiferromagnetic dimer condensation in TlCuCl3 and BaCuSi2O6. Clarifying a quantum critical point in BaCuSi2O6 and spontaneous electric polarization in TlCuCl3 upon entering the magnon condensate phase were the subjects of recent investigation. These findings imply that condensate management may be possible when an electric field is introduced. Microwave pulses may be used to create non-equilibrium magnon condensates, which allow the chemical potential to be controlled regardless of temperature. A significant magnon density is produced at a critical pumping power, allowing it to thermalize and condense. Using light-scattering spectroscopy, this was initially seen in thin films of yttrium iron garnet (YIG), a ferrimagnet with a long spin-lattice relaxation time that permits magnon condensation at room temperature and is effectively pumped. A room-temperature magnon supercurrent a macroscopic collective motion of magnon condensate exposed to a thermal gradient was discovered more recently via research on YIG films. In the context of spintronic devices operating at ambient temperature, this finding is intriguing.

Exciton and exciton-polariton condensates. When it comes to the on-demand management of macroscopic coherent states of matter, indirect exciton and exciton-polariton condensates produced by optical excitation represent the state-of-the-art. In GaAs quantum wells, coupled layers of two-dimensional electron gas (2DEG) continue to be the gold standard for studying these condensates. Van der Waals materials have great potential for producing designer condensates, including graphene and TMDs. Hexagonal boron nitride (hBN) insulating spacers allow for layer-by-layer tailoring of the 2DEG interlayer spacing. The gating control of the intralayer doping is complemented by this handy tuning knob. High exciton binding energies, light effective masses, and the importance of many-body effects in TMD-based heterostructures suggest that excitonic condensates and superfluids might be realized under ambient conditions. In the quantum Hall regime, data for two bilayer graphene specimens separated by a small hBN spacer provide the tantalizing glimpses of exciton condensation. In this system, indirect excitons develop between partly filled Landau

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levels rather than requiring optical activation (Fig. 3c). Exciting findings with microcavity exciton-polariton condensates are still being obtained.

Revealing hidden phases and new states of matter

A common feature of several families of quantum materials is insulator-to-metal transitions (IMT). Investigating these transitions in the realms of ultrahigh electric/magnetic fields, in conjunction with ultrafast and intense laser pulses and nanoscale spatial resolution, is now possible. These hitherto unachievable methods of control and investigation have revealed a variety of metastable states and "hidden" phases that are not visible in traditional phase diagrams of quantum materials, which are usually recorded with area-averaging probes at equilibrium.

Anomalous metallicity at the nanoscale. First, we consider the veteran quantum material V2O3, which is well known as a model Mott insulator. However, this molecule keeps finding new aspects of relationships. A structural alteration is concomitant with the electrical transition at TIMT~160 K. In addition, the low-T insulator exhibits antiferromagnetic ordering consistent with Mott theory predictions. Figure 4a's nano-infrared picture illustrates the mechanics of the first-order phase transition. One may understand the scattering amplitude data in terms of local conductivity101 by looking at the picture and the histogram plot in the bottom panel. These results show that insulating (blue) and metallic (red) domains coexist. These characteristics apply to all other associated oxides undergoing the IMT. The ongoing change in V2O3's local infrared conductivity across the IMT is an unexpected outcome. This result contradicts the traditional model of a first-order transition, in which the two phases stay electrically constant during the transition and the metallic volume percentage rises with temperature at the cost of the insulating volume fraction. Since the discovery required a scanning probe experiment with nanoscale precision, the observed continuous feature of the IMT remained elusive, even though it was anticipated within a model that allowed for a long-range interaction between the domains.

Phononic controls of quantum materials. Light-induced metallicity of the correlated oxide Pr0.7Ca0.03MnO3, which is caused by a bandgap collapse by coherent excitation of Mn–O vibrational modes, is a well-known early example. Resonant phonon excitation reduces lattice and electron heating, two undesirable consequences that are difficult to prevent when materials are pumped non-discriminatively with accessible near-IR or visible lasers. Phononic excitation has been used to melt orbital order and produce significant effective magnetic fields in addition to generating metallicity. Phononically induced enhancement of superconducting correlations in the cuprates and in K3C60 has been observed in a particularly interesting set of studies. The actual component of the optical conductivity for K3C60 at 100 K, which is much higher than the compound's bulk Tc, is shown in Figure 4d. A Drude-like reaction is shown at equilibrium. When 0.18 eV pulses are used for excitation, a gap reminiscent of superconductivity emerges. These results have spurred theoretical efforts to clarify the potential source of the superconducting amplification and to comprehend the nature of lattice dynamics resulting from nonlinear phonon excitation. With respect to the average boson occupancy and the on-site Coulomb potential U, the theoretical phase diagram shown in the inset of Fig. 4d predicts improved superconductivity in addition to additional phases.

Need for attosecond transients. The need to separate the electronic and lattice components during light-activated processes is a basic issue in ultra-fast research. Obviously, the electrical processes are far quicker than the lattice dynamics, which are hard to investigate with ordinary lasers that have pulse lengths of just a few tens of femtoseconds. Because of this, the light-induced band gap decrease in VO2 that was seen in reference 120 was described as a "instantaneous" process. We see that the ability of attosecond pump–probe studies to differentiate between these ostensibly instantaneous electrical events and delayed femto–second processes affecting the lattice is growing. Therefore, a wider application of attosecond methods to quantum materials is expected to provide important breakthroughs. Attosecond pulses also allow for damage-free exposure of insu- lating solids to electric fields on the order of the Zener critical field Fcrit = Eg/ea, which is an extra benefit of this area of study. It was shown that wide-bandgap dielectrics' a.c. conductivity may be increased by more than 18 orders of magnitude in less than a millisecond by using ultra-rapid reversible optical waveforms. These studies pave the way for signal processing in the petahertz range, which is beyond the capabilities of standard electronics.

Topological phenomena under control

According to the Landau paradigm, symmetries identify different phases of matter. There is, nonetheless, a more precise categorization system that is based on the topological or entanglement characteristics of the electronic wavefunction rather than symmetry. The latter result in unusual static and finite-frequency response functions as well as exotic phases of matter with protected states that are caused by geometric (Berry) phase effects.26,123–126. This section focuses on methods for controlling Berry phases associated with either linear or quadratic band crossings of the bulk electronic structure in order to achieve topological events on demand. Since there are already many great articles on interacting topological phases, including topological superconductors, which are a rapidly developing field of study with possible implications to quantum computing, we won't get into that here.

By closing the gap in the bulk Dirac spectrum and reopening it with reversed orbital character, this family of materials may be created from trivial insulators. By using impurity doping, pressure/strain, temperature, heterostructuring/ electronstatic-gating, or even optical stimulation, this kind of trivial-to-topological insulator transition may be accomplished and provide helical edge/surface states with non-trivial Berry phase on demand.

Hall (Chern) anomalous quantum insulators. Quantum anomalous Hall insulators are created via spontaneous time-reversal symmetry breaking to open a gap in the 2D surface Dirac spectrum of a 3D topological insulator and tailoring the Fermi level to reside within the surface gap134. This non-trivial phase does not need an externally applied magnetic field and shows a quantized Hall conductance e2/h similar to typical quantum Hall insulators. In gated ferromagnetic three-dimensional topological insulator Cry(BixSb1−x)2−yTe3 devices, it has been experimentally shown that a quantum anomalous Hall phase135 develops below a Curie temperature TC = 15 K, where the chiral modes can be turned on and off by altering the temperature across TC.

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