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Perspective on multiple degrees of freedom in crystal materials

SHI GuoQiang & XUE DongFeng*

Multiscale Crystal Materials Research Center, Shenzhen Institute of Advanced Technology, Chinese Academy of Sciences, Shenzhen 518055, China

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Degrees of freedom describe the freedom of variables or values to vary within a specific material system. Research on functional crystalline materials generally involves their preparation and growth to large-sized crystals, as well as the exploration of structure-property relationships [1,2]. With the continuous research on the applications of functional crystalline materials in quantum science and technology, the problem of multiple degrees of freedom is garnering increasing attention [3,4]. Notably, crystalline materials possess multiple degrees of freedom, such as those of the lattice, charge, spin, orbital, and topology of electronic states, as shown in Figure 1 [3]. Furthermore, a certain degree of freedom that undergoes a generalized order-disorder transition near the phase-transition point is known as the order parameter. Materials with multiple order parameters and strong interactions are called quantum materials. Typically, quantum materials are regarded as functional materials that possess energy origins beyond their electronic contributions. With the development of quantum technology, the application of crystalline materials continues to attract increasing attention [5–10]. The strong coupling among the various degrees of freedom in crystalline materials is key to their application as quantum materials. To meet the high requirements of quantum technology applications, crystalline materials with multiple quantum degrees of freedom that exhibit strong coupling are required.

The coupling among the various degrees of freedom in crystalline materials can be realized through external perturbations [3,11]. For example, when the physical dimension of a material is the same as the characteristic length of quantum particles (i.e., the exciton Bohr radius or mean free path), the quantum particles in the material will exhibit controlled spin entanglement arising from the quantum-limited nature of the domain. Furthermore, under external effects such as high pressure, the crystal structure will distort, causing atoms to deviate from their regular lattice equilibrium positions, resulting in lattice distortion, electron-lattice coupling, and orbital hybridization and rehybridization [7]. Lattice vibrations (phonons) change the potential distribution within a crystal, resulting in a change in the electronic state, followed by electron-phonon coupling. Notably, electron-phonon coupling is a mechanism for conventional superconductivity [12]. Strong many-body interactions in crystalline materials can also lead to the coupling of multiple degrees of freedom; these include electron-electron, exciton-exciton, and spin-valley interactions. Many-body interactions induce collective behavior, giving rise to magnetic and charge-density waves. Spin-valley interactions occur in monolayer two-dimensional transition metal chalcogenides, causing an optical rotation effect and the valley Hall effect [13]. Unusual phenomena in topological insulators can also be explored by coupling multiple degrees of freedom [9]. The spin Hall effect is the effect of the spin-orbit coupling of electrons. The most well-known two-dimensional topological insulator is the quantum spin Hall insulator. Under an external electric field, electrons with upward and downward spins generate two Hall currents in opposite directions. These currents cancel each other, re-

*Corresponding author (email: df.xue@siat.ac.cn)

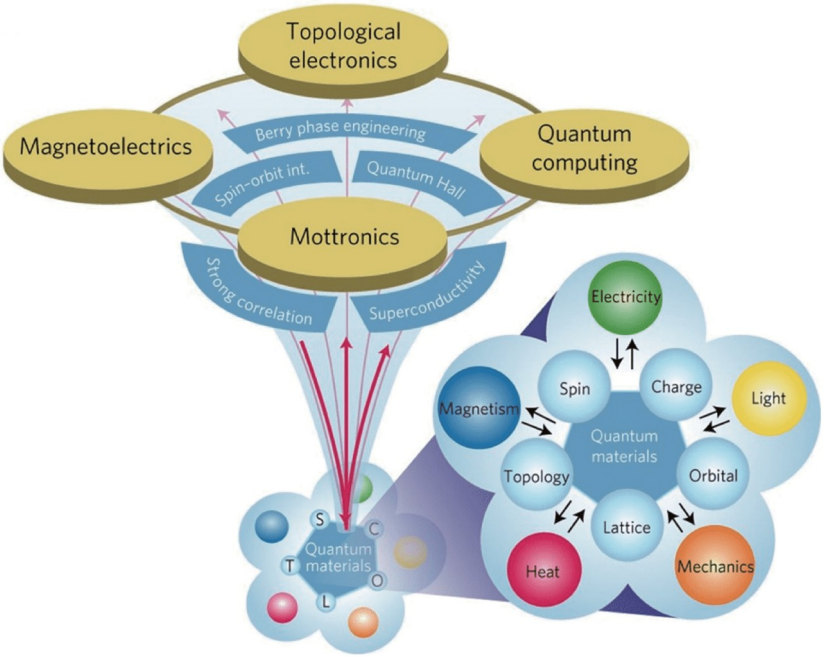

Figure 1 (Color online) Coupling among degrees of freedom in quantum materials results in many intriguing phenomena under different external fields [3]. Copyright@2017, Springer Nature.

sulting in a net-zero Hall conductance. However, a quantized spin Hall conductance remains owing to the spin current.

In correlated electronic systems, such as systems containing rare-earth ions, electronic interactions can be regulated by parameters other than temperature, such as pressure, magnetic field, and doping, thereby gradually suppressing long-range magnetic ordering and inducing quantum phase transitions [14–16]. Substantial quantum critical fluctuations near quantum critical points can profoundly affect the physical properties at finite temperatures and induce exotic quantum states or quantum phenomena such as unconventional superconductivity and non-Fermi-liquid behavior. For example, the electronic structure of $CeRh_6Ge_4$ evolves with increasing temperature. The 4f and conduction-band electrons exhibit an apparent hybridization behavior above the ferromagnetic transition temperature over a wide temperature range. The 4f electronic states have significantly different temperature dependences at other momentum points. Additionally, the hybridization strength of the 4f and conduction-band electrons has pronounced anisotropy. Specifically, the hybridization strength in the direction along the Ce atomic chain is significantly higher than that in the vertical direction, which may be related to the *quasi*-one-dimensional arrangement of the 4f magnetic moment. This anisotropic hybridization of Ce 4f electrons may be essential for generating ferromagnetic quantum criticalities.

Lithium niobate (LN) single-crystals have essential applications in optical quantum communication because of the coupling of multiple degrees of freedom [17–20]. LN single-crystals have multiple degrees of freedom, including lattice, charge, and orbital degrees of freedom. The coupling of multiple degrees of freedom can be achieved through multiscale quantum design for quantum applications. Electronegativity (EN) strongly affects orbital hybridization. The orbital hybridization of Li−O and Nb−O yields Li_xO_y and Nb_uO_w clusters, respectively (Figure 2). From the atomic/ionic scale to the mesoscale, EN may serve as a degree of freedom. Furthermore, the chemical bonds within the clusters become soft, and the ions are rearranged in three dimensions. During orbital rehybridization, the chemical bonds become stiff, thereby forming the lattice. Intrinsic defects are formed during the crystallization process. The chemical environments (such as vacancy population and mass conservation) of atoms or defects may serve as constraints that can decrease the degree of freedom at a given site. Intrinsic defects induce the rehybridization of valence electron orbitals, which results in symmetry breaking and a change in the local energy domain (Figure 2). Concurrently, domain engineering enables the functional optimization of LN. The existence of an external field leads to defect-lattice and electron-phonon couplings in the LN structure. Therefore, it is crucial to construct LN-based quantum systems that combine multiple degrees of freedom in LN single crystals through multiscale concepts and to study the possibility of the coupling of multiple degrees of freedom.

Research on crystalline materials with multiple degrees of freedom is of great practical significance for advancing the applications of such materials in quantum technology. Increasing the lifetime of quantum states through materials design and control technologies based on external light,

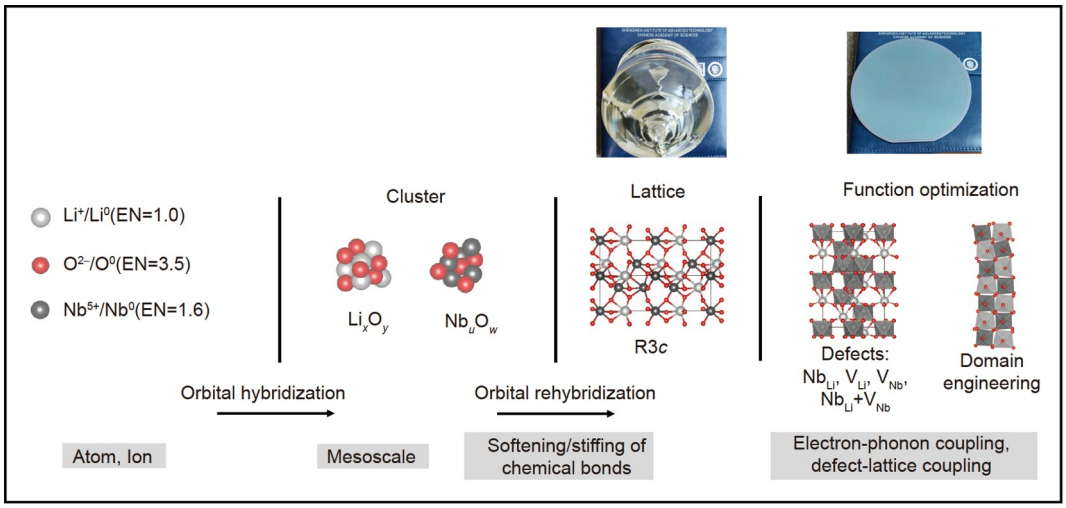

Figure 2 (Color online) Multiple-degree-of-freedom coupling processes in the crystal structure of LN. Based on a multiscale process, Li, Nb, and O form clusters through the hybridization of valence electron orbitals. Simultaneously, the system undergoes a dynamic mesoscale process, and the clusters are rehybridized to form a lattice. After lattice formation, the multiple degrees of freedom, including the charge and lattice degrees of freedom, in the LN single crystal structure are coupled to achieve functional optimization. EN stands for electronegativity. Softening of chemical bonds indicates decreased bond strength, while stiffening of chemical bonds indicates increased bond strength. The defects symbols on the right-hand side of the figure are defined as follows: Nb_{Li}:Nb antisite, V_{Li}:Li vacancy, V_{Nb}:Nb vacancy, and Nb_{Li} + V_{Nb}:Nb antisite and Nb vacancy.

electricity, magnetic fields, and pressure is one application of quantum materials. The crucial factor is the multiple degrees of freedom in crystalline materials and their coupling effects. The challenges and prospects of current research in this area are as follows.

(1) Introducing degrees of freedom, such valleys, in crystalline materials to create strongly correlated electronic systems.

(2) Realizing multiple-degree-of-freedom coupling processes in crystalline materials, such as the coupling of degrees of freedom through external fields, including ultrafast laser-induced many-body interactions.

(3) Inducing the transition of the degrees of freedom in a crystalline material at the phase-transition point (order parameter), symmetry breaking in the structure, and constructing quantum systems.

(4) Investigating rare-earth ions as promising candidates for solid-state quantum memory devices because of the long-lived superposition states of their optical and spin transitions.

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