extending static DFT to its molecular dynamics finite temperature limit.

It turned out that the imposed symmetry constraints constituted a self-fulfilling prophecy, being a key reason for the incorrect predictions. This limitation was not part of the DFT method itself, representing instead a naive application of the DFT method. Removing the artificially imposed high symmetry resulted in lowering the total energy (meaning, stabilization), simultaneously converting false metals into real Mott insulators even without strong correlation.

The simplest case of long-range-ordered quantum phases at $T < T_{(LRO)}$

Row 1 in Fig. 1 illustrates long-range-ordered (LRO) periodic organization of local lattice motifs that typically occur below the ordering temperatures, $T < T_{(LRO)}$. Such local motifs include patterns of atomic displacements (1a in Fig. 1), magnetic moments (1b in Fig. 1) or dipole moments (1c in Fig. 1), and are commonly observed experimentally as crystallographic, magnetic or ferroelectric long-range order. Using such observed organization of local lattice motifs as input to DFT (or optimizing them via total energy minimization) generally produces a good description of the metal versus insulator phenomena at $T < T_{(LRO)}$. Examples of predictions of such true insulators replacing the false metals found from naive DFT include cuprate superconductors[14].

The mechanism for gapping depends on the dominant symmetry-breaking modes, be that atomic distortion in $LaMnO_3$ (1a in Fig. 1), magnetic moment ordering in antiferromagnets $LaTiO_3$ and $SrMnO_3$ (1b in Fig. 1), or ferroelectric dipoles (1c in Fig. 1). If, however, the magnitude symmetry breaking is too weak and insufficient to open the gap, the result of DFT is a failed Mott insulator, or simply a 'true metal' (for example, $SrVO_3$). Thus, symmetry-broken DFT generally works well unless one artificially imposes a highly symmetric periodic cell where symmetry breaking is geometrically excluded, even if it were to lower the total energy. This would then result in a false metal instead of a real insulator, irrespective of the DFT exchange correlation used.

Para-phases above the $T > T_{(LRO)}$

Unlike the LRO ground-state structures of row 1 in Fig. 1, para-phases (rows 2 or 3 in Fig. 1) appearing at higher temperatures lack long-range order (but they can have correlated disorder and short-range order). They can appear as paraelastic (2a in Fig. 1), paramagnetic (2b in Fig. 1) or paraelectric (2c in Fig. 1). Much like a chemically disordered A_xB_{1-x} alloy that also lacks long-range order, in all of these cases the cell size and cell-internal motifs needed for their description in band theory are generally unknown. Such para-phases were simplistically imagined to be made of single motifs ('monomorphous' in row 3 in Fig. 1),

as gleaned also from characterization techniques that delivered the global averaged structure. Band-structure calculations for such monomorphous high-symmetry structures generally predicted a (false) metallic state for systems with an odd number of electrons per cell[10,11,14]. In retrospect, this practice of using the average configuration, washing out all local motifs, as input to electronic structure calculations misdirected the field.

It turned out that such an imposed symmetry constraint was the key factor for the false-metal predictions in para-phases of Mott insulators. Significantly, the calculated total energy of such high-symmetry 'virtual crystal', symmetry-unbroken para-phases[15–18] were predicted to be 1–2 eV higher than the symmetry-broken magnetic cases, pointing to the fact that such symmetry-unbroken models (including refs. [11,12]) are irrelevant competing phases.

Recent developments

Research in the DFT community pointed out that there are avenues for removing the constraints on such naive DFT[15–19] rather than disposing of DFT altogether. Considering larger-than-minimal unit cells instead revealed a significant lowering of the total energy by breaking the symmetry of the assumed ideal configurations, simultaneously converting paramagnets ($LaTiO_3$, $LaMnO_3$, $SrMnO_3$ and NiO) from false metals into real insulators even without strong correlation. The rise of the insulating gap relative to its approximate false metal reference is often a result of different forms of symmetry breaking. The fall of the insulating gap in forming a metallic phase (insulator to metal transition) is often the result of the weakening or elimination of symmetry breaking by temperature or pressure.

On the experimental side, increasing use of local probes that do not average over large volumes has meant that it is possible to 'see' the local positional, magnetic and dipolar configurations. Recent observations[20,21] have reported that the nominally cubic paraelectric oxide phases of $BaTiO_3$ and $KNbO_3$ are piezoelectric, suggesting that it is cubic only as a global average, but not locally.

It has also been recently noted that in several materials that were traditionally believed to be controlled primarily by strong interelectronic physics manifesting 'electronic phase of matter', including Mott insulators and paramagnetic nickelates, "the lattice in fact plays a crucial role"[22].

So where has all the strong correlation gone?

mean-field-like DFT and by (2) explicitly
highly correlated symmetry-preserving
methods beg the obvious question: what
happened to the strong correlation when
symmetry was broken in a larger cell? Indeed,
correlation is representation dependent
rather than being an absolute statement on
the intrinsic physical nature of a compound
or a phase, as we have been conditioned
to think. What is strong correlation in