The expanding condensed matter multiverse

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Condensed matter physicists sometimes pity our colleagues in high-energy physics. They are limited to studying a single vacuum and its excitations: the particles of the standard model. For condensed matter physicists every new phase of matter brings a new 'vacuum'. Remarkably the low-energy excitations of these new vacua can be very different from the individual electrons, protons and neutrons that constitute the material. The condensed matter multiverse contains universes where the particle-like excitations carry only a fraction of the elementary electronic charge (*1*), are magnetic monopoles (*2*), or are their own antiparticles (*3*). None of these properties have ever been observed in the particles found in free space. Often emergent gauge fields accompany these 'fractionalized' particles (*2, 4, 5*), just as electromagnetic gauge fields accompany charged particles. On page XXX of this issue, Hassan et al. (*6*) provide a glimpse of the emergent behaviors of a putative new phase of matter – the dipole-liquid. But, what particles live in this universe and what new physics is found in this and neighboring parts of the multiverse?

Crystalline solids are easy to distinguish from fluids – liquids and gases look the same everywhere, but the periodic arrays of atoms in crystals break translational and rotational symmetries. This broken symmetry leads directly to the important differences between a crystal and a fluid, for example the crystal's rigidity (*7*). The differences between gases and liquids are more subtle. Particles move freely in a gas, rarely interacting with one another, like cars on the open road; in a liquid the motion of particles is correlated, i.e., depends on what other particles are doing, like city driving (see figure). Continuing this analogy a glass is like a traffic jam, interactions between particles seem to bring movement to a halt, and a crystal resembles a parking lot with every particle neatly in place.

Some phases of matter are easier to detect than others. In ferromagnets the spins of unpaired electrons (radicals) align, creating a net magnetization (see figure). This allowed ancient civilizations to discover ferromagnetism. In antiferromagnets neighboring spins point in opposite directions leaving no net magnetic moment. Consequently, antiferromagnetism was not observed until the twentieth century. We still await conclusive experimental evidence for the existence of topological spin-liquids – proposed phases where the spins lack long-range order, but display long-range quantum correlations known as entanglement (*5*).

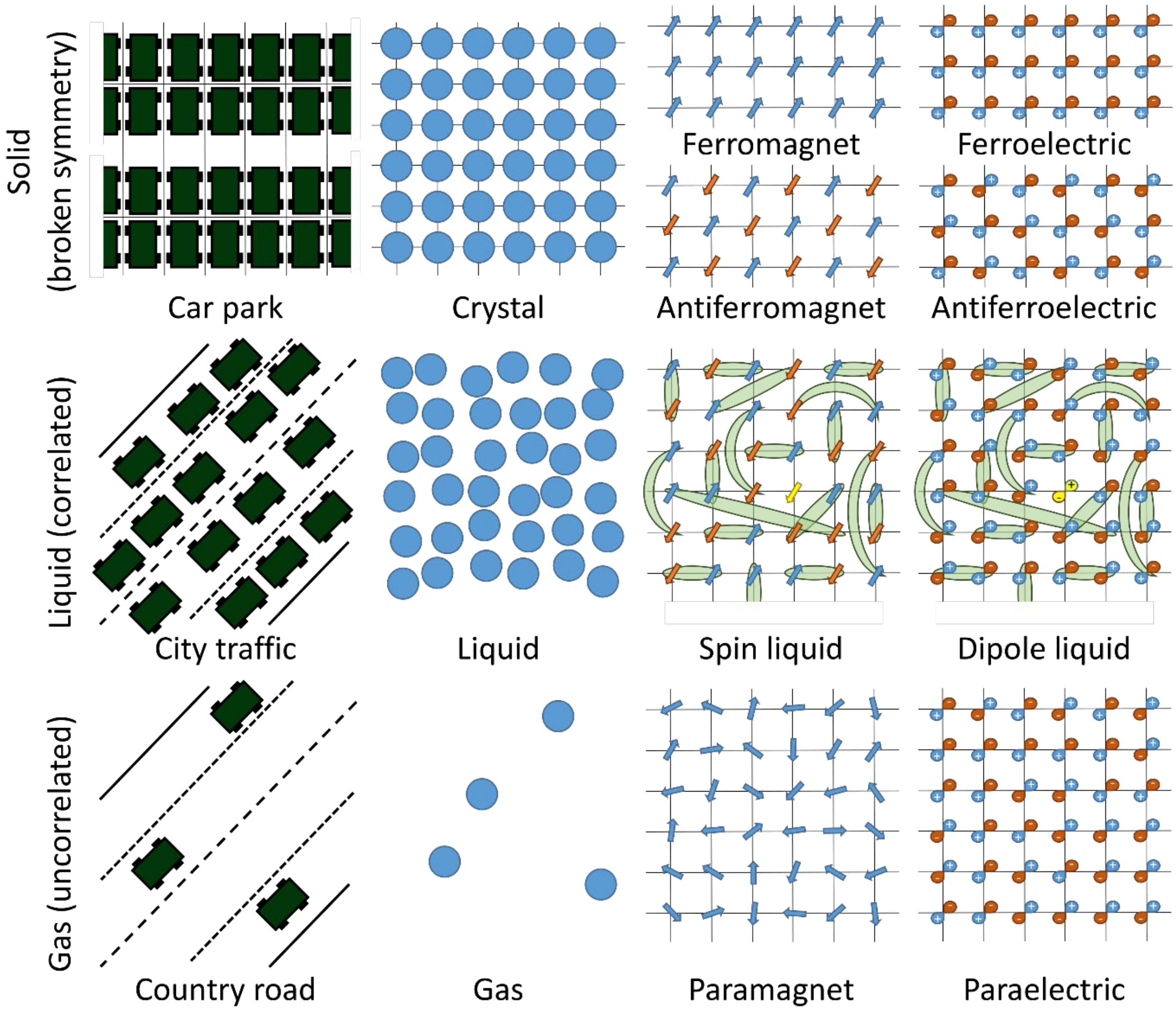
Ferroelectrics and antiferroelectrics are phases where electric dipoles align parallel and antiparallel to their nearest neighbors (see figure). Simple paramagnets and paraelectrics, with little correlation between spins or dipoles respectively, are essentially spin- or dipole-gases. (Anti)ferromagnets and (anti)ferroelectrics could be described as spin- and dipole-crystals, as a symmetry is broken. Similarly, systems with strong correlations between spins or dipoles but no broken symmetry, are known as spin- (*4, 5*) and dipole-liquids (*6, 8, 9*).

Hassan et al. exploit two special features of molecular crystals. The $(BEDT\text{-}TTF)_2$ salts they study are dimer Mott insulators: most $(BEDT\text{-}TTF)_2$ dimers carry a charge of +1. Excitations that increase the charge of one dimer are bound to excitations that decreases the charge on a nearby dimer, just as electrons are bound to atomic nuclei. Classically the positive charge resides on one molecule or other, hence there are two states that differ by the flip of an electric dipole. Quantum mechanics allows tunneling between these two states, as in the Creutz-Taube ion (*10*). If the dipole tunnels quickly compared to other relevant time scales then there is a quantum superposition of the two classical states and we have a dipole gas. Hassan et al. use Raman scattering to probe the behaviors of two vibrational modes of the molecules – one sensitive to the charge on the molecule, the other insensitive. This enables them to study the dipolar fluctuations.

κ-$(BEDT\text{-}TTF)_2Hg(SCN)_2Cl$ is a dipole solid at low temperatures, spontaneously breaking the inversion symmetry of the $(BEDT\text{-}TTF)_2^+$ dimers. However, in κ-$(BEDT\text{-}TTF)_2Hg(SCN)_2Br$ the dipoles fluctuate rapidly, leading to a dipole fluid. The first piece of evidence that this is a dipole-liquid rather than a dipole gas is the observation of a low energy continuum of excitations in the Raman spectrum. Secondly, the low temperature heat capacity of the dipole solid in κ-$(BEDT\text{-}TTF)_2Hg(SCN)_2Cl$ is proportional to temperature cubed, as one would expect from phonons or other bosonic excitations. But, in κ-$(BEDT\text{-}TTF)_2Hg(SCN)_2Br$ there is a linear term in the heat capacity. A natural source of a linear term in the heat capacity is excitations that are itinerant fermions. Hassan et al. argue that both of these experiments are witnessing either collective excitations of the dipoles or hybrid spin-dipole excitations driven by interactions between the dipoles and the unpaired spins of the $(BEDT\text{-}TTF)_2^+$ radicals. In a broken symmetry phase, such as a dipole solid, the collective excitations are (Goldstone) bosons (*7*). But in a dipole-liquid the excitations might fractionalize into fermionic particles, as appears to happen in some spin-liquids (*4, 5*) (see figure).

If the excitations have hybrid spin-dipole character, might spin-dipole interactions also drive the exotic physics of the BEDT-TTF salts (*11*)? Little is known about the spin physics in κ-$(BEDT\text{-}TTF)_2Hg(SCN)_2Br$, so Hassan et al. turn to κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$, which is believed to be a spin-liquid (*4*). They find no evidence of dipole-liquid or -solid phases in this material, suggesting that its physics arises solely from the spins.

Like many of the most interesting experiments, Hassan et al.'s work contains more questions than answers. Hassan et al. have discovered an exciting new universe ripe for exploration. Conclusively demonstrating that the excitations to its 'vacuum' are itinerant fermions may require new experimental tools. BEDT-TTF salts often become superconducting under pressure (*4*). Many physicists believe that this superconductivity results from the material wanting, but failing, to become a spin-liquid (*4, 5*). Do failed dipole-liquids also superconduct? How common are dipole-liquids? There is evidence for one in $BaFe_{12}O_{19}$ (*8*) and proposals for building them from polar molecules (*9*). Might the quantum behaviors of ice X (*12*) be understood as a dipole-liquid?



Solid, liquid and gaseous phases of particles, spins and dipoles and their automotive analogies. In crystalline solids a symmetry is broken by the long-range order. In gases the correlations between the constituents are weak. Liquids are intermediate with strong correlations between the constituents but no long-range order. In the sketches of spin and dipole-liquids the green curves indicate quantum correlations between spins/dipoles pointing in opposite directions. One possible route to liquid phases is that both spins/dipoles flip simultaneously, this is an example of quantum entanglement. A quantum state composed of many configurations similar to that sketched above can produce a spin-liquid (*4, 5*). Excitations in such a state involve unpaired spins (highlighted in yellow) that move about the crystal – spinons. Depending on the details of the quantum state the spinons can obey either Fermi or Bose statistics (*4, 5*). The sketch for the dipole-liquid is speculative. The suggestion, implicit in Hassan et al.'s work, that the physics of dipole-liquids is similar to that of spin-liquids and can support itinerant fermionic quasiparticles, remains to be substantiated.

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