Utilizing Chemical Intuition in the Search for New Quantum Materials

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Antiferromagnetism and topologically non-trivial properties joined in EuSn$_2$P$_2$, a compound by design.

Building on the technological achievements of the 20th century, driven by semiconductor technology and miniaturization, the 21st century has evolved an unquenchable thirst for new materials that exhibit exotic properties to master the grand challenges of today, including smart electronic devices, quantum computing, and efficient catalysts for sustainable energy conversion. The specialization of such systems calls for strategies to design tailor-made materials from scratch to satisfy increasingly complex requirements. In this very issue of ACS Central Science, Gui et al. take the reader on an inspiring journey utilizing rational design to synthesize a new topological insulator candidate that exhibits intrinsic long-range magnetic order.¹

So far, scientists that aimed for unconventional quantum states originating from “topology+” used the design of heterostructures to combine different electronic phases from distinct materials, rather than realizing such phenomena within a single material.²,³

Almost a decade later, Gui et al. have now taken up the challenge to design an AFM-TI and deduced some general design rules from a chemist’s perspective:¹ First, they emphasize the importance of choosing elements that are known to establish different kinds of chemical bonding and...
exhibit large spin–orbit coupling to access the mentioned band inversion. Second, they point out that known unconventional materials (Bi2Se3, Cd3As2, WTe2, etc.) are often low-dimensional in their electronic structure, thus avoiding the interference of other electronic bands with the topological feature of interest. Third, in search for magnetic properties, the incorporation of an element whose magnetic properties stem from localized, hence weakly interacting, $4f$ electrons, was necessary. Linking all of these ideas toward a new material is ambitious, but this is exactly what the authors have achieved in their work by designing and realizing the novel compound EuSn$_2$P$_2$.

The layered EuSn$_2$P$_2$ (cf. Figure 1) is a new member of the group of two-dimensional solids, a hallmark of which is a low-dimensional electronic structure. In the stacking direction, 2D materials are generally weakly bound and exhibit a clear cleavage plane that is beneficial for studying the surface physics mentioned above. Additionally, this compound adds to the small subgroup of magnetic 2D materials. EuSn$_2$P$_2$ is antiferromagnetic, since the magnetic moments of Eu$^{2+}$ ions align ferromagnetically in a layer, oriented perpendicular to the stacking direction, but couple in the opposite direction compared to the neighboring layers. Furthermore, EuSn$_2$P$_2$ is straightforward to synthesize for a trained solid-state chemist and its magnetic properties already emerge below 30 K, an experimentally well-accessible temperature, making the compound a prime candidate for future experiments. Gui et al. employ the common suite of solid-state characterization techniques to thoroughly describe the magnetism, transport properties, and electronic band structure. The material is weakly metallic and exhibits a large concentration of holes responsible for the transport of electronic charge, which in turn is sensitive to the intrinsic magnetic state.

At this point, it is instructive to look at the electronic band structure close to the Fermi level up to which the bands are filled with electrons. This is where interesting topological physics can emerge, for example, in the form of protected band crossings, multiply degenerate points, or band inversions, which may give rise to exotic surface states or host quasiparticles such as Dirac or Majorana fermions. The experimental signature of such peculiarities in the band structure—is they are not occluded by topologically trivial features—is an unusual behavior in magneto-transport measurements.

The authors used comprehensive DFT calculations and indeed identified the band inversion of a topological insulator in the band structure. A real crystal (unlike an ideal one having an infinitely periodic crystal structure) is terminated at its surface; in the case of a layered material, this is most likely the 2D cleavage plane. Considering different possible terminations (Eu, Sn, or P), the authors calculated the shape of the characteristic surface states. In the lab, the method of choice to record the electronic structure at a single crystal’s surface is angle-resolved photoemission spectroscopy (ARPES). Only quantum oscillations can be more telling about the Fermi surface, but this method requires an even better single crystal quality than ARPES. With ARPES, the authors were able to reveal a close agreement between experimental and calculated electronic bands and, by the shape of the surface states, they were able to conclude that the
termination of the crystal is phosphorus. Finally, they digged deep into theoretical condensed-matter physics and calculated the $Z_2$ invariant (a classification scheme for topological insulators based on symmetry), confirming $\text{EuSn}_3\text{P}_2$ to be an antiferromagnetic, strong topological insulator.

The experimental realization of an AFM-TI has implications for fundamental research into exotic phenomena such as quantized magnetoelectric coupling, intrinsic axion insulator state, chiral Majorana fermions, or the quantum anomalous Hall effect. These prospects have already resonated with the scientific community, and just recently, Otrokov and co-workers succeeded in the synthesis of another AFM-TI material, MnBi$_2$Te$_3$, a van der Waals compound with out-of-plane FM layers. And yet, a major drag on the rapidly evolving field of topological materials is the small number of experimentally confirmed materials. For the field to flourish, realistic candidates are desperately needed that are easy to fabricate as large single crystals, do not display structural over-complexity due to large-scale atomic disorder or vacancies, and exhibit the electronic transitions of interest at accessible temperatures. By training, chemists are equipped with the unique ability to synthesize new compounds, to swiftly evaluate crystal structures based on their symmetry, and to routinely draw on well-developed bonding theories based on size relations or electronegativity of the elements. 200 years of empirical knowledge have bestowed chemists with a certain intuition for thermodynamic stability, reactivity, and crystal chemistry.

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To summarize, Gui et al. succeeded in the rational design and realization of a new magnetic quantum material, $\text{EuSn}_3\text{P}_2$. This accomplishment serves as an inspiration for collaborations across disciplines and encourages a wider community of chemists to get involved. The significant strides made in this work toward a new family of exciting materials by marrying topologically nontrivial properties with magnetism are only the beginning. It will be interesting to see what is next.

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Funding
We gratefully acknowledge financial support by the Max Planck Society.

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