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Uncovering hidden local states in a quantum material

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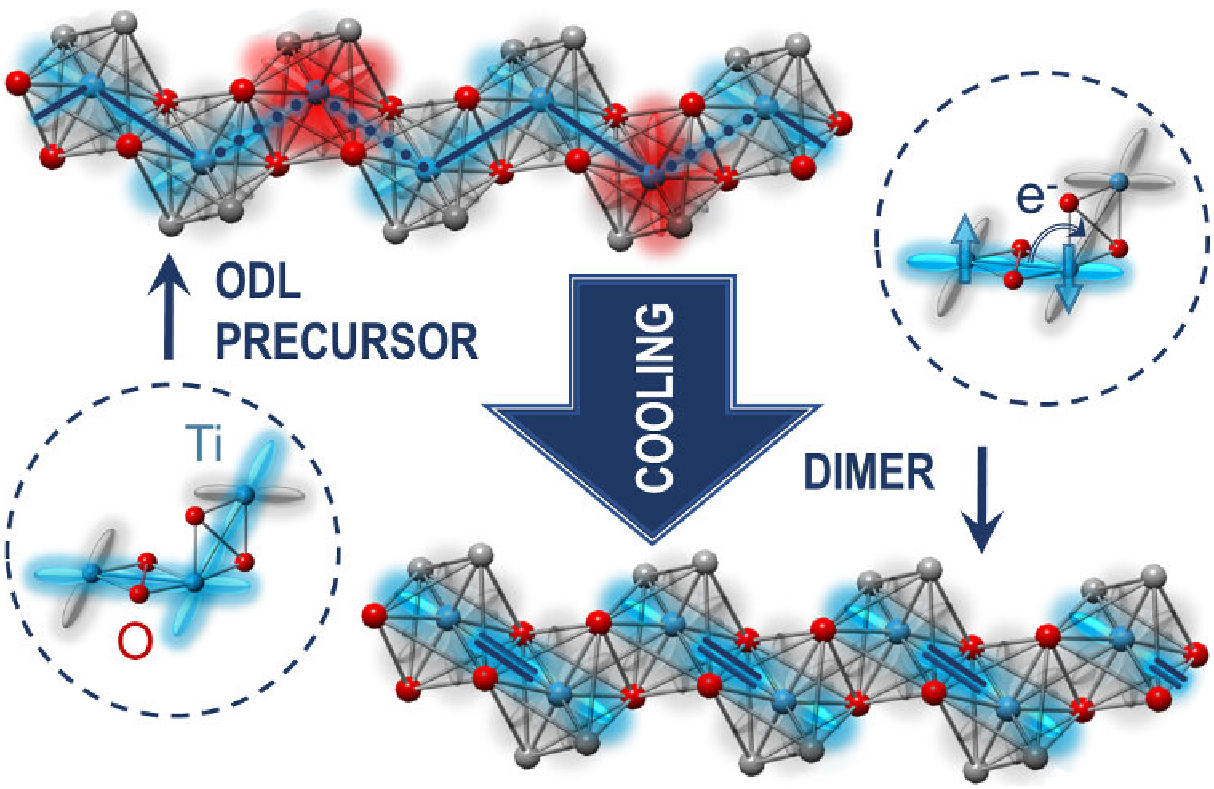

Scientists have collected evidence of local symmetry breaking in a quantum material upon heating. They believe these local states are associated with electronic orbitals that serve as orbital degeneracy lifting (ODL) "precursors" to the titanium (Ti) dimers (two molecules linked together) formed when the material is cooled to low temperature. (Electron orbitals are considered degenerate when they have the same energy levels.) Understanding the role of these ODL precursors may offer scientists a path forward in designing materials with the desired technologically relevant properties, which typically emerge at low temperature. Credit: Brookhaven National Laboratory

Quantum materials display exotic behaviors due to the effects of quantum mechanics, or how matter acts on the very small scale of atoms and subatomic particles. The technologically relevant properties of quantum materials result from complex interactions of electron charge, orbital, and spin and their coupling to the material's crystal structure. For example, in some materials, electrons can flow freely without any resistance; this phenomenon, called superconductivity, could be harnessed to transmit power more efficiently. Typically, these properties emerge at low temperature, where crystals display low (broken) structural symmetry.

"Not surprisingly, this low-temperature regime is well studied," said Emil Bozin, a physicist in the X-ray Scattering Group of the Condensed Matter Physics and Materials Science (CMPMS) Division at the U.S. Department of Energy's (DOE) Brookhaven National Laboratory. "Meanwhile, the high-temperature regime remains largely unexplored because it's associated with relatively high symmetry, which is considered uninteresting."

But Bozin and colleagues have recently discovered states of local symmetry breaking at high temperature. These local states are associated with electronic orbitals (regions within an atom where electrons are most likely to be found) that serve as orbital degeneracy lifting (ODL) "precursors" to what happens at low temperature. Orbital degeneracy refers to when orbitals have the same energy. Lifting of this degeneracy means that some orbitals will have a relatively higher energy and others a lower energy.

"We think such local states are in some way enablers of the material properties of interest that materialize at much-lower temperature," explained Bozin.

The scientists first observed these local states in 2019 in a material (copper iridium sulfide) with a metal-insulator transition and in an iron-based superconductor. Now, the team—representing Brookhaven Lab; DOE's Oak Ridge National Laboratory; University of Tennessee, Knoxville; and Columbia University—has found them in an insulator containing sodium, titanium, silicon, and oxygen. This insulating material is one of the minerals forming the Earth's upper mantle. Beyond the geological interest, it's a candidate for quantum spin liquids (QSLs), an exotic state of matter in which electron spins remain fluid-like to the lowest temperatures, constantly fluctuating. QSLs could provide a material platform for quantum computing, spintronics (electronics based on electron spin rather than charge), superconductivity, and other technologies.

"Our findings suggest that this ODL precursor behavior at high temperature may be quite common and should be considered in theoretical studies to understand the functional properties of quantum materials," said physicist Weiguo Yin of the CMPMS Division Condensed Matter Theory Group.

To probe the material's atomic structure, the team analyzed how the material scattered neutrons and X-rays. Both probes are needed because of their different sensitivities to particular elements based on atomic weight. Unlike X-rays, neutrons can distinguish light elements, like oxygen. With the neutron and X-ray scattering patterns, the local arrangement of atoms can be deduced through the atomic pair distribution function (PDF), which describes the distances between different atoms in a sample. Using software, scientists can then find the structural model that best fits the experimental atomic PDF function.

Their analysis revealed signatures of local symmetry breaking far above the temperature at which the material undergoes a structural transition to form titanium dimers (two molecules linked together). When the material is heated, these dimers seem to disappear, but really, they stick around, evolving into a dual ODL state.

"The high-temperature, high-crystallographic-symmetry state assumes the presence of orbital degeneracy, but orbital degeneracy may not be energetically favorable," said Bozin. "As we see here, the dimers get replaced, and what remains is a locally distorted crystal structure. This distortion lifts the degeneracy of two orbitals and allows the system to enter a lower-energy state."

Next, the team plans to tailor orbital properties in this material—for example, by switching out titanium with ruthenium, which will change the electron count and is predicted to provide a better QSL. They will also see whether the ODL precursors exist in other materials and how they are related to phenomena of interest, such as superconductivity. In particular, they would like to explore systems with different degrees of spin-orbit coupling, which is an alternative mechanism for ODL.

"The discovery of these orbital precursors helps us to better understand the competition between different low-temperature quantum states—an understanding that will allow us to tilt the playing field to get materials with desired low-temperature properties," said Simon Billinge, a physicist in the CMPMS Division X-ray Scattering Group and professor of materials science and engineering and of applied physics and mathematics at Columbia University.

More information: R. J. Koch et al, Dual Orbital Degeneracy Lifting in a Strongly Correlated Electron System, *Physical Review Letters* (2021). DOI: 10.1103/PhysRevLett.126.186402

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