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Seeking Moments of Disorder

The future of technology relies, to a great extent, on new materials, but the work of developing those materials begins years before any specific application for them is known. Stephen Wilson, a professor of materials in UC Santa Barbara's College of Engineering, works in that "long before" realm, seeking to create new materials that exhibit desirable new states.

In the paper "Field-tunable quantum disordered ground state in the triangular-lattice antiferromagnet NaYbO2," published in the journal Nature Physics, Wilson and colleagues Leon Balents, of the campus's Kavli Institute for Theoretical Physics, and Mark Sherwin, a professor in the Department of Physics, describe their discovery of a long-sought "quantum spin liquid state" in the material NaYbO2 (sodium ytterbium oxide). The study was led by materials student Mitchell Bordelon and also involved physics students Chunxiao Liu, Marzieh Kavand and Yuanqi Lyu, and undergraduate chemistry student Lorenzo Posthuma, as well as collaborators at Boston College and at the U.S. National Institute of Standards and Technology.

At the atomic level, electrons in one material's lattice structure behave differently, both individually and collectively, from those in another material. Specifically, the "spin," or the electron's intrinsic magnetic moment (akin to an innate bar magnet) and its tendency to communicate and coordinate with the magnetic moments of nearby electrons differs by material. Various types of spin systems and collective patterns of ordering of these moments are known to occur, and materials scientists are ever seeking new ones, including those that have been hypothesized but not yet shown to exist.

“There are certain, more classical moments that let you know to a very high degree of certainty that the spin is pointing in a particular direction,” Wilson explained. “In those, the quantum effects are small. But there are certain moments where the quantum effects are large, and you can’t precisely orient the spin, so there is uncertainty, which we call ‘quantum fluctuation.’”

Quantum magnetic states are those in which the magnetism of a material is primarily driven by such quantum fluctuations, generally derived from the uncertainty principle, intrinsic to magnetic moments. “So, you envision a magnetic moment, but the uncertainty principle says that I can’t perfectly orient that in any one direction,” Wilson noted.

Explaining the quantum spin liquid state, which was proposed long ago and is the subject of this paper, Wilson said, “In conventional materials, the magnetic moments talk to one another and want to orient relative to one another to form some pattern of order.” In classical materials, this order is disrupted by thermal fluctuations, what Wilson describes as “just heat from the environment.”

“If the material is warm enough, it is nonmagnetic, meaning the moments are all sort of jumbled relative to one another,” he explained. “Once the material is cooled, the moments start to communicate, such that their connection to one another outcompetes the thermal fluctuations and they form an ordered state. That’s classical magnetism.”

But things are different in the quantum world, and magnetic moments that fluctuate can actually be the inherent “ground state” of a material.

“So, you can ask if there is a magnetic state in which the moments are precluded from freezing or forming some pattern of long-range order relative to one another, not by thermal fluctuations, but instead, by quantum fluctuations,” Wilson said. “Quantum fluctuations become more relevant as a material cools, while thermal fluctuations increase as it heats up, so you want to find a magnet that doesn’t order until you can get it cool enough such that the quantum fluctuations preclude it from ever ordering.”

That quantum disorder is desirable because it is associated with entanglement, the quantum mechanical quality that makes it possible to encode quantum information. To determine whether NaYbO2 might exhibit that characteristic, the researchers had to determine the intrinsic, or ground state of the material’s magnetic moments when all thermal fluctuations are removed. In this particular system, Wilson was able to determine experimentally that the magnetic moments are intrinsically in a fluctuating, disordered state, thus confirming that a quantum disordered state exists.

To find the hypothesized state, said Wilson, “First you have to put highly quantum magnetic moments into a material, but your material needs to be constructed such that the moments don’t want to order. You do that by using the principle of ‘magnetic frustration.’”

A simple way to think of that, according to Wilson, is to imagine a single triangle in the lattice structure of the material. “Let’s say I build my material so that the magnetic moments are all located on a triangular lattice,” he said, “and they all talk to one another in a way that has them wanting to orient antiferromagnetically, or antiparallel, to one another.”

In that arrangement, any adjacent moment on the triangle wants to orient antiparallel to its neighbor. But because there are an odd number of points, you have one up at one point and one down (antiparallel to the first) at the second point, meaning that the third moment has a differently oriented moment on each side, so it doesn’t know what to do. All of the moments are competing with one another.

“That’s magnetic frustration, and, as it turns out, it reduces the temperature at which the moments are finally able to find some arrangement they all agree on,” Wilson said. “So, for instance, classically, nature decides that at some temperature the mismatched moments agree that they will all point to 120 degrees relative to each other. So they’re not all 100 percent happy but it’s some compromise that establishes an ordered state.”

From there, he added, “The idea is to take a frustrated lattice where you have already suppressed the ordered state, and add quantum fluctuations to it, which take over as you cool the material. Magnetic frustration lowers the ordering temperature enough so that quantum fluctuations eventually take over and the system can stabilize into a fundamentally disordered quantum spin state.”

Wilson continued: “That’s the paradigm of what people are looking for; however, some materials may seem to display this state when actually, they don’t. For instance, all real materials have disorder, such as chemical or structural disorder, and this can also prevent the magnetic moments from talking to each other effectively and becoming ordered. In such a case, Wilson says, “They might form a disordered state, but it’s more of a frozen, or static, disordered state than it is a dynamic quantum state.

“So, if I have a magnetic system that doesn’t order at the lowest temperatures I can measure, it can be tricky trying to understand whether what I’m measuring is an intrinsic quantum spin liquid fluctuating type of state or a frozen, extrinsic, chemically driven disordered state. That is always debated.”

Among the most interesting findings about this new material, Wilson said, is that even at the lowest measurable temperature — .005 degree Centigrade above absolute zero — it still doesn’t order.

“However, in this material we can also apply a magnetic field, which breaks this competition engendered by magnetic frustration, and then we can drive it to order, inducing a special kind of antiferromagnetic state,” he added. “The reason that’s important is because this special state is very delicate and a very good fingerprint for how much chemical disorder there is in the system and its influence on the magnetic ground state. The fact that we can drive this field-driven state tells us that the disordered state we see at low temperature with zero magnetic field is indeed an intrinsically quantum disordered state, consistent with being a quantum spin liquid state.”

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