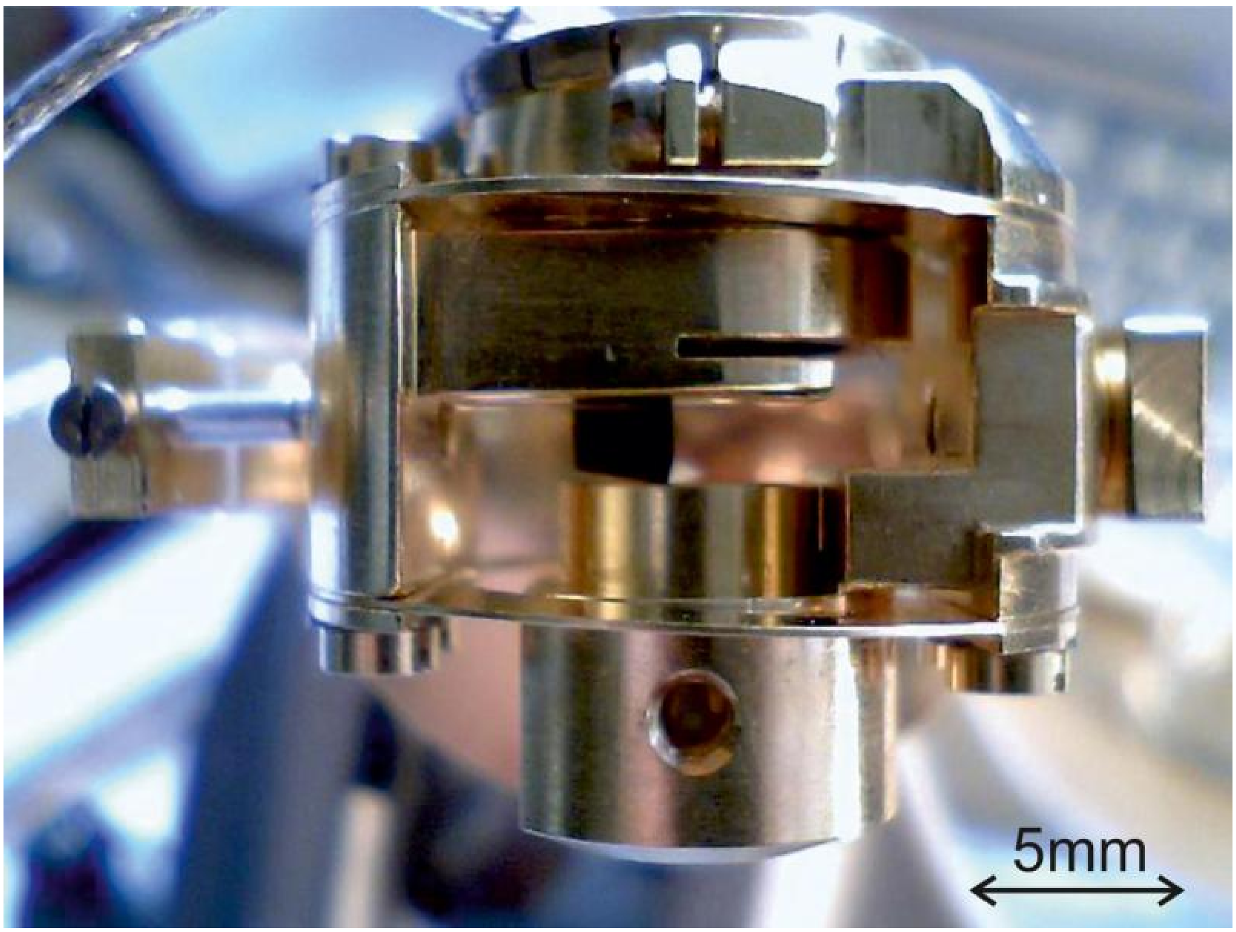
Entropy landscape sheds light on quantum mystery

May 12 2017



Physicists at Karlsruhe Institute of Technology used this capacitive dilatometer to measure the thermal expansion in cerium copper gold alloys cooled to temperatures very close to absolute zero with a precision of one tenth of a trillionth of a meter, or approximately one-thousandth the radius of single atom. The precise thermal expansion measurements allowed the researchers to map out the stress dependence of entropy in materials as they were cooled to the point of a quantum phase transition. Credit: K. Grube/Karlsruhe Institute of Technology

By precisely measuring the entropy of a cerium copper gold alloy with baffling electronic properties cooled to nearly absolute zero, physicists in Germany and the United States have gleaned new evidence about the possible causes of high-temperature superconductivity and similar phenomena.

"This demonstration provides a foundation to better understand how novel behaviors like high-temperature superconductivity are brought about when certain kinds of materials are cooled to a quantum critical point," said Rice University physicist Qimiao Si, co-author of a new study about the research in this week's *Nature Physics*.

The experimental research was led by Hilbert von Löhneysen of the Karlsruhe Institute of Technology in Karlsruhe, Germany. Löhneysen's team, including study lead author Kai Grube, spent a year conducting dozens of experiments on a compound made of cerium copper and gold. By studying the effect of stress, or pressure applied in specific directions, and by making the materials very cold, the team subtly changed the spacing between the atoms in the crystalline metallic compounds and thus altered their electronic properties.

The cerium copper gold alloys are "heavy fermions," one of several of types of quantum materials that exhibit exotic electronic properties when very cold. The best-known of these are high-temperature superconductors, so named for their ability to conduct electrical current with zero resistance at temperatures well above those of traditional superconductors. Heavy fermions exhibit a different oddity: Their electrons appear to be effectively hundreds of times more massive than normal and, equally unusual, the effective electron mass seems to vary strongly as temperature changes.

These odd behaviors defy traditional physical theories. They also occur at very cold temperatures and come about when the materials are tuned to a "quantum phase transition"—a change from one state to another, like ice melting. In 2001, Si and colleagues offered a new theory: At the quantum critical point, electrons fluctuate between two entirely different quantum states, so much so that their effective mass becomes infinitely large. The theory predicted certain tell-tale signs as the quantum critical point is approached, and Si has worked with experimental physicists for the past 16 years to amass evidence to support the theory.

"Liquid water and ice are two of the classical states in which H2O can exist," said Si, director of the Rice Center for Quantum Materials. "Ice is a very ordered phase because the H2O molecules are neatly arranged in a crystal lattice. Water is less ordered compared with ice, but flowing water molecules still have underlying order. The critical point is where things are fluctuating between these two types of order. It's the point where H2O molecules sort of want to go to the pattern according to ice and sort of want to go to the pattern according to water.

"It's very similar in a quantum phase transition," he said. "Even though this transition is driven by quantum mechanics, it is still a critical point where there's maximum fluctuation between two ordered states. In this case, the fluctuations are related to the ordering of the 'spins' of electrons in the material."

Spin is an inherent property—like eye color—and every electron's spin is classified as being either "up" or "down." In magnets, like iron, spins are aligned in the same direction. But many materials exhibit the opposite behavior: Their spins alternate in a repeating up, down, up, down pattern that physicists refer to as "antiferromagnetic."

Hundreds of experiments on heavy fermions, high-temperature superconductors and other quantum materials have found that magnetic order differs on either side of a quantum critical point. Typically, experiments find antiferromagnetic order in one range of chemical composition, and a new state of order on the other side of the critical point.

"A reasonable picture is that you can have an antiferromagnetic order of spins, where the spins are quite ordered, and you can have another state in which the spins are less ordered," said Si, Rice's Harry C. and Olga K. Wiess Professor of Physics and Astronomy. "The critical point is where fluctuations between these two states are at their maximum."

The cerium copper gold compound has become a prototype heavy fermion material for quantum criticality, largely due to the work of von Löhneysen's group.

"In 2000, we did inelastic neutron scattering experiments in the quantum critical cerium copper gold system," said von Löhneysen. "We found a spatial-temporal profile so unusual that it could not be understood in terms of the standard theory of metal."

Si said that study was one of the important factors that stimulated him and his co-authors to offer their 2001 theory, which helped explain von Löhneysen's puzzling results. In subsequent studies, Si and colleagues also predicted that entropy—a classical thermodynamic property—would increase as quantum fluctuations increased near a quantum critical point. The well-documented properties of cerium copper gold provided a unique opportunity to test the theory, Si said.

In cerium copper-six, substituting small amounts of gold for copper allows researchers to slightly increase the spacing between atoms. In the critical composition, the alloys undergo an antiferromagnetic quantum phase transition. By studying this composition and measuring the entropy numerous times under varying conditions of stress, the Karlsruhe team was able to create a 3-D map that showed how entropy at very low yet finite temperature steadily increased as the system approached the quantum critical point.

No direct measure of entropy exists, but the ratio of entropy changes to stress is directly proportional to another ratio that can be measured: the amount the sample expands or contracts due to changes in temperature. To enable the measurements at the extraordinarily low temperatures required, the Karlsruhe team developed a method for accurately measuring length changes of less than one tenth of a trillionth of a meter—approximately one-thousandth the radius of a single atom.

"We measured the entropy as a function of stress applied along all the different principal directions," said Grube, a senior researcher at Karlsruhe Institute of Technology. "We made a detailed map of the entropy landscape in the multidimensional parameter space and verified that the quantum critical point sits on top of the entropy mountain."

Von Löhneysen said the thermodynamic measurements also provide new insights into the quantum fluctuations near the critical point.

"Surprisingly, this methodology allows us to reconstruct the underlying spatial profile of quantum critical fluctuations in this quantum critical material," he said. "This is the first time that this kind of methodology has been applied."

Si said it came as a surprise that this could be done using nothing more than entropy measurements.

"It is quite remarkable that the entropy landscape can connect so well with the detailed profile of the quantum critical fluctuations determined from microscopic experiments such as inelastic neutron scattering, all the more so when both end up providing direct evidence to support the theory," he said.

More generally, the demonstration of the pronounced entropy enhancement at a quantum critical point in a multidimensional parameter space raises new insights into the way electron-electron interactions give rise to high-temperature superconductivity, Si said.

"One way to relieve the accumulated entropy of a quantum critical point is for the electrons in the system to reorganize themselves into novel phases," he said. "Among the possible phases that ensue is unconventional superconductivity, in which the electrons pair up and form a coherent macroscopic quantum state."

More information: K. Grube et al. Multidimensional entropy landscape of quantum criticality, *Nature Physics* (2017). DOI: 10.1038/nphys4113

Provided by Rice University