Surprising electronic disorder in a copper oxide-based ceramic

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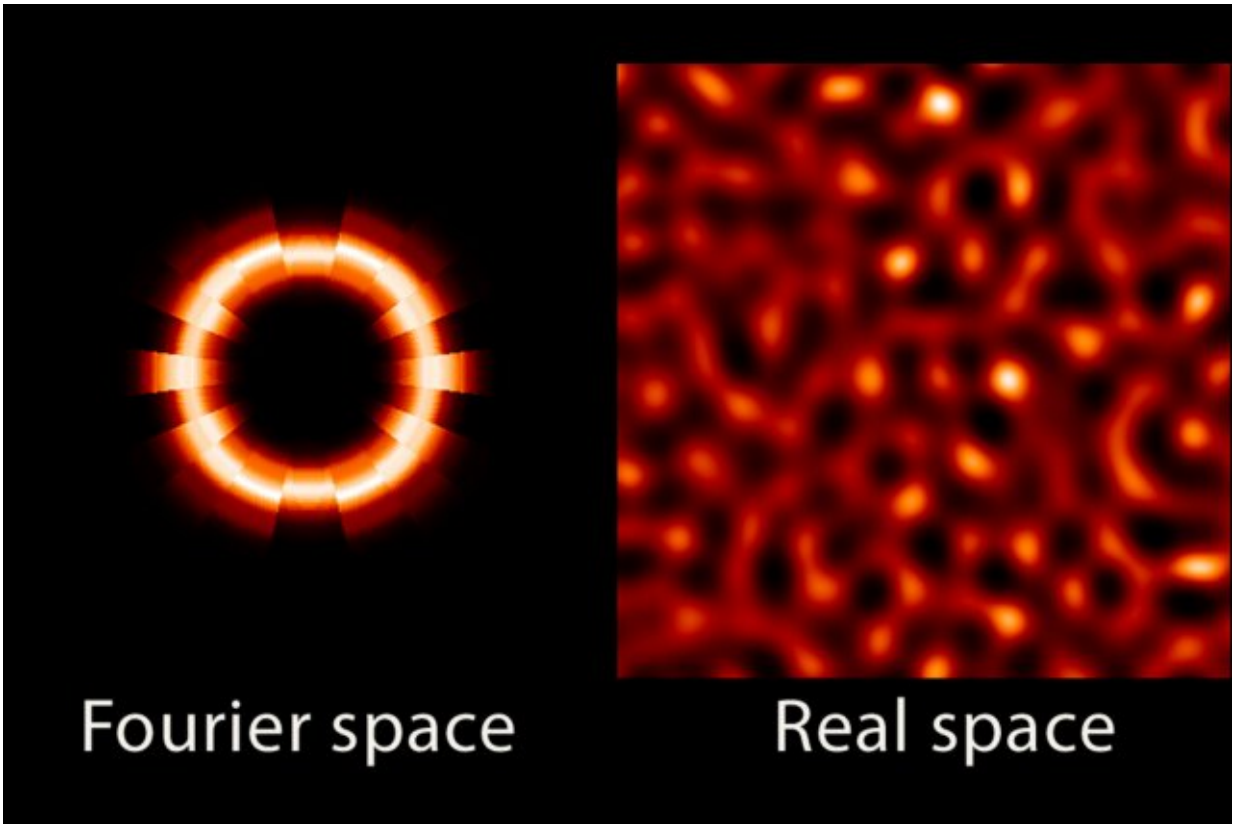

Experiments on ultra-thin copper-oxide ceramic superconductors performed by MIT researchers showed an unexpected disorderly, or "glassy," arrangement of electrons, known as a "Wigner glass." At left, "Fourier space," or momentum space, shows the diffraction data that proved the tendency of the charge ripples to align in any direction, while the image at right displays the random placement of electrons in "real space." Credit: Min Gu Kang

Cuprates, a class of copper-oxide ceramics that share a common building block of copper and oxygen atoms in a flat square lattice, have been studied for their ability to be superconducting at relatively high temperatures. In their pristine state, however, they are a special kind of insulator (a material that does not readily conduct electricity) known as a Mott insulator.

When electrical charge carriers—either electrons or the lack of electrons, known as "holes"—are added to an insulator in a process called doping, the insulator may become a metal, which readily conducts electricity, or a semiconductor, which can conduct electricity depending on the environment. Cuprates, however, behave neither like a normal insulator nor like a normal metal because of strong interactions between their electrons. To avoid the large energy cost arising from these interactions, the electrons spontaneously organize in a collective state where the motion of each particle is tied to all the other ones.

One example is the superconducting state, where electrons move in unison and drift with zero net friction when a potential is applied, a zero-resistance state which is a defining characteristic of a superconductor. Another collective electronic state is a "charge density wave," a term coined from the wave-like modulation in the density of electrons, in which electrons "freeze" into periodic and static patterns, at the same time hindering electron flow. This state is antagonistic to the superconducting state, and, therefore, important to study and understand. In cuprates, charge-density-waves prefer to align to the atomic rows of copper and oxygen atoms that make up the underlying crystal structure, with wave "'crests" occurring every three to five unit cells, depending on the material and doping level.

Using a technique known as resonant X-ray scattering to study these charge-density-waves in two different cuprate compounds, neodymium copper oxide (Nd_2CuO_4 or NCO) and praseodymium copper oxide

(Pr_2CuO_4 or PCO) doped with extra electrons, MIT researchers made an unexpected discovery. Their work revealed a phase of the material where the electrons fall into a disordered, or "glassy," arrangement, dubbed a "Wigner glass." The results were recently published in a paper in *Nature Physics*.

Resonant X-ray scattering is a recently developed diffraction technique in which crystallography is performed on electrons rather than exclusively on the atoms as in conventional X-ray diffraction. "In the limit of low concentration of doped electrons, we observed a completely new and unexpected form of electronic phase which is neither a superfluid nor a crystal, but it rather has the characteristics of a Wigner glass. In this phase, the electrons form a collective state without any orientational preference," says the paper's senior author Riccardo Comin, assistant professor of physics at MIT. Such an amorphous glass of electrons is completely unprecedented in this family of materials, he adds.

This phenomenon emerges only in a narrow window of electron doping. "Intriguingly, this exotic new state only exists in a small region of the electronic phase diagram of this material, and when more electrons are doped in the [copper oxide] planes, a more conventional electronic crystal is recovered, whose ripples align to the crystallographic axes of the underlying atomic lattice," Min Gu Kang, the paper's lead author, explains.

The MIT team, consisting of Comin, graduate student Kang, and postdoc Jonathan Pelliciari, designed the project and led the majority of experiments. Their research was made possible by the contributions of researchers at various institutions and facilities worldwide. Resonant X-ray scattering measurements were performed at multiple synchrotron facilities including the Berlin Electron Storage Ring in Germany, the Canadian Light Source in Saskatoon, Saskatchewan, Canada, and the

Advanced Light Source, in Berkeley, California. The copper-oxide thin film samples were grown at NTT Basic Research Laboratories in Japan. Theoretical analysis was developed by researchers at the Indian Institute of Science in India.

Comin notes that the proposed theory explains the role of the electronic band structure in governing the periodic spacing and lack of orientational preference of the density waves as a function of doping level in this material. "Our theory suggests that these electronic ripples are initially formed with irregular shapes and are likely nucleated around defects or impurities in the material," Comin says. "When the density of carriers increases, the electrons manage to find a more highly-ordered arrangement that minimizes the total energy of the system, thereby restoring the more conventional charge density waves that have been observed universally in all families of copper-oxide superconductors."

"I was completely blown away by Riccardo's results on NCO and PCO," says Peter Abbamonte, Fox Family Professor in Engineering at the University of Illinois at Urbana-Champaign, who developed the resonant soft X-ray scattering technique. Noting that charge density wave (CDW) order in cuprates has been at the center of the field for well over a decade, Abbamonte, who was not involved in this research, explains that the previous understanding has been that the CDW order is pinned to the crystal lattice, meaning the charge density wave must point in either of two perpendicular directions, but nowhere in between. This conventional wisdom is built on two decades of resonant scattering and scanning tunneling microscopy experiments that have always found this to be the case, he notes.

Comin's research on these particular electron-doped cuprates showed that during the glassy phase the charge order can point in any direction, independent of the crystal lattice it lives in. "The more precise statement is that the CDW order parameter is not Ising-like (that is, taking only

discrete values, in this case two: x or y), as has always been assumed, but is more like an X-Y order parameter (that is, free to choose any value on a continuous range, such as all directions between x and y as is the case here) that is only weakly influenced by the crystal," Abbamonte says.

"It is going to take some time for the community to fully digest this realization and its implications for understanding the relevance of CDW order," Abbamonte adds. "What is clear is Riccardo's paper is going to lead to a serious re-reckoning of the rules of the game, and in this sense is a major advance for the field."

Superconductors have an immense, largely untapped potential for transformative applications such as quantum computing, lossless energy transport, magnetic sensing and medical diagnostic imaging, and plasma and nuclear fusion power technologies.

"Overall, our study has revealed yet another manifestation of the exquisite quantum character of charge carriers in high-temperature superconductors, which ultimately arises from the nature of the electronic interactions," Comin says. "The detailed behavior of electrons uncovered in this work provides new insights on how high-temperature superconductivity is born out of a Mott insulator, and promises to bridge a gap between regions of the phase diagram with very contrasting phenomenologies."

More information: Mingu Kang et al. Evolution of charge order topology across a magnetic phase transition in cuprate superconductors, *Nature Physics* (2019). DOI: 10.1038/s41567-018-0401-8