Nanoscale technique uses atomic vibrations to show how quantum materials behave at interfaces

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With the SSTS technique, a laser pulse is applied to one side of an oxide/metal sample and terahertz radiation is emitted. From the signal, the dynamics of the TO1 surface phonon is detected in the oxide at its interface with the metal. Credit: Argonne National Laboratory

Scientists are racing to develop new materials for quantum technologies in computing and sensing for ultraprecise measurements. For these future technologies to transition from the laboratory to real-world applications, a much deeper understanding is needed of the behavior near surfaces, especially those at interfaces between materials.

Scientists at the U.S. Department of Energy's (DOE) Argonne National Laboratory have unveiled a new technique that could help advance the development of quantum technology. Their innovation, surface-sensitive spintronic terahertz spectroscopy (SSTS), provides an unprecedented look at how quantum materials behave at interfaces.

The work is published in the journal *Science Advances*.

"This technique allows us to study surface phonons—the collective vibrations of atoms at a material's surface or interface between materials," said Zhaodong Chu, a postdoctoral researcher at Argonne and first author of the study. "Our findings reveal striking differences between surface phonons and those in the bulk material, opening new avenues for research and applications."

In materials such as crystals, atoms form repeating patterns called lattices, which can vibrate in waves known as phonons. While much is understood about phonons in the bulk material, little is known about surface phonons—those occurring within nanometers of an interface. The team's research reveals that surface phonons behave differently, enabling unique quantum behaviors such as interfacial superconductivity.

Superconductivity, the phenomenon of electrons flowing without resistance, has applications in technologies such as MRI machines and particle accelerators. Interfacial superconductivity—a type that appears only at the boundary between two materials—holds promise for new quantum technologies.

"The idea for this discovery began with the finding some years ago that interfaces between two crystalline materials can exhibit superconducting behavior neither one shows on its own," said Anand Bhattacharya, an Argonne physicist.

"It is only when the two materials are together that the superconductivity magic happens at the interface, which is different from the bulk," added Argonne physicist Haidan Wen.

Believing that a specific type of vibration in the crystal—called the TO1 phonon—triggers this interfacial superconductivity, the team set out to find direct evidence of its role.

There were two main challenges, Wen explained. First, the interface is buried in the sample and only a few nanometers thick, making it hard to study using conventional methods. Second, the team needed to work with terahertz radiation. This happens in a frequency range a thousand times higher than 5G phone networks. Many important quantum effects happen in this terahertz range but capturing them with high resolution is difficult.

The researchers used their SSTS method on samples made by depositing a thin magnetic film onto an oxide crystal. In this method, ultrafast laser pulses pass through the oxide crystal and strike the thin magnetic layer. The interaction between laser light and matter then produces terahertz vibrations at the oxide interface.

By using this technique, the team detected the TO1 phonon. They also showed that the phonon's behavior within 5 nanometers of the interface differed from the bulk. Surface phonons are like waves in the shallow end of a lake—they behave differently than those in deeper waters.

"Our interface-sensitive technique can be applied to a broad range of materials for probing elusive quantum behavior, including magnetism and superconductivity," said Michael Norman, Argonne Distinguished Fellow and director of the Argonne Quantum Institute. "We now have a new window into quantum materials that can point the way to novel quantum devices for future technologies."

Bhattacharya added, "Terahertz light interacting with matter can not only probe quantum materials in new ways, as in our study, but also induce entirely new states of matter. This is an incredibly exciting avenue for future investigation."

In addition to those quoted above, Argonne authors include Junyi Yang, Yan Li, Jianguo Wen, Ashley Bielinski, Qi Zhang, Alex Martinson, Stephan Hruszkewycz and Dillon Fong. Also contributing were Xiaodong Xu and Kyle Hwangbo from the University of Washington.

More information: Zhaodong Chu et al, Revealing subterahertz atomic vibrations in quantum paraelectrics by surface-sensitive spintronic terahertz spectroscopy, *Science Advances* (2024). DOI: 10.1126/sciadv.ads8601

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