Pines' Demon Observation at Sr_2RuO_4

Discovery of strange metal behavior predicted 67 years ago

1. Overview

In 1956, the US theoretical physicist David Pines predicted a strange state of electrons in solids. Normally, electrons have mass and charge, but Pines believed that electrons could combine to form composite particles that were massless, electrically neutral, and did not interact with light. He named this new particle "DEM-on", as an acronym for "particle responsible for the distinct electron motion". However, until now this particle has never actually been observed.

A research team led by Professor Peter Abbamonte of the University of Illinois at Urbana-Champaign (UIUC), USA, and Professor Yoshiteru Maeno of the Toyoda-Riken-Kyoto University Collaboration (TRiKUC), Kyoto University Advanced Research Institute, as a member, has finally discovered it, 67 years after the Pines' demon was predicted. As described in a paper recently published in Nature, the team observed the behaviour of the demon in strontium-ruthenium oxide using a special technique that directly excites the electronic mode of the material. "Demons have been theoretically conjectured for a long time, but no experiments have been reported. In fact, we were not initially searching for that state, but it turned out that the novel electronic excitation mode we observed was in fact the one predicted by Pines."

One of the most important discoveries in condensed matter physics is that electrons often lose their individuality in solids. Through electrical interactions, electrons may combine to form collective units. With sufficient energy, electrons can even form new composite particles with charge and mass determined by electrical interactions, well known as "plasmons". However, the mass is usually so large that plasmons cannot be created as spontaneous thermal excitations at room temperature.

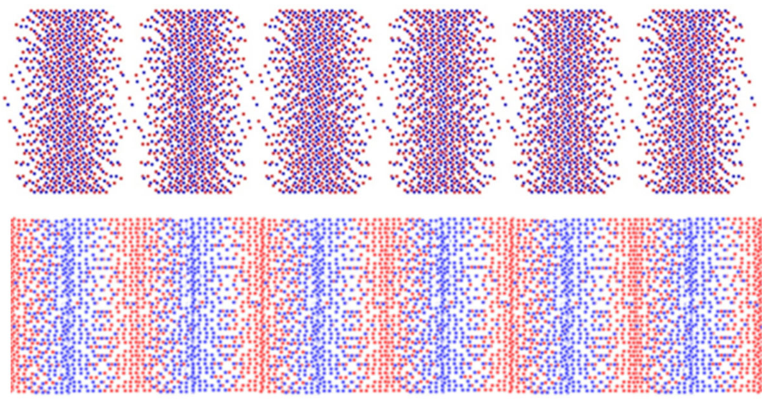

Figure 1. Oscillatory electron densities for (top) ordinary plasmons and (bottom) Pines' demons. Electrons in the β (beta) and γ (gamma) bands are depicted in red and blue.

Pines predicted an exception. If electrons in a solid have multiple energy bands, as many metals do, he argued, each plasmon can combine in an out-of-phase pattern to form a new plasmon, or demon, which is massless and neutral (Fig. 1). As demons are massless, they can form at any energy and can exist with thermal excitation at any temperature.

This has led to speculation that demons may have an important influence on the behaviour of multiband metals with multiple electronic bands. Demons are electrically neutral and may not leave traces in experiments with simple materials.

2． Research methods and results

The research team included Dr. Chanchal Sow, Postdoctoral Research Fellow, Graduate School of Science, Kyoto University (at the time of the research, now Associate Professor, Indian Institute of Technology Kanpur), Professor Yoshiteru Maeno of the Toyoda Riken - Kyoto University Research Centre (TRiKUC), Kyoto University Institute for Advanced Study, (at the start of the research, Professor of the Graduate School of Science at the same university), Ali A. Husain, Edwin W. Huang, Melinda S. Rak, Samantha I. Rubeck, Xuefei Guo, Tai C. Chiang, Professor Philip W. Phillips and Professor Peter Abbamonte of the University of Illinois, USA, Matteo Mitrano, Harvard University, USA; Hongbin Yang and Philip E. Batson, Rutgers University, USA; and Bruno Uchoa, University of Oklahoma, USA.

A new excitation mode has been observed in momentum-resolved electron energy loss spectroscopy (M-EELS) using crystals of the strontium-ruthenium oxide Sr_2RuO_4 grown at Kyoto University. Unlike the well-known plasmon, it is long-wavelength and gapless, and can be interpreted as a "Pines' demon", which is an oscillation in the number of electron occupancies between bands rather than an oscillation in electron density. 1956 Pines was the quantum responsible for Distinct Electron Motion (DEM) "DEM-on" (Demon), and this is the first reported observation of this mode.

The new mode observed in Sr_2RuO_4 with M-EELS (Fig. 2) is confirmed to be a Pines' demon by its gapless behaviour and the momentum dependence of its intensity and critical momentum. The theoretically predicted demons are electrically neutral excited modes consisting of out-of-phase motion of the electron occupancy numbers in the β and γ bands, which are acoustic modes with zero energy gap in the long wavelength limit and cannot be excited by light.

Demons are thought to be important in various phenomena, including phase transitions in mixed-valence semimetals, optical properties of metal nanoparticles, and high-temperature superconductivity in metal hydrides. Our work fulfills a prediction made 67 years ago, and it is expected that the Pines' demon is in fact widespread in other multiband metals.

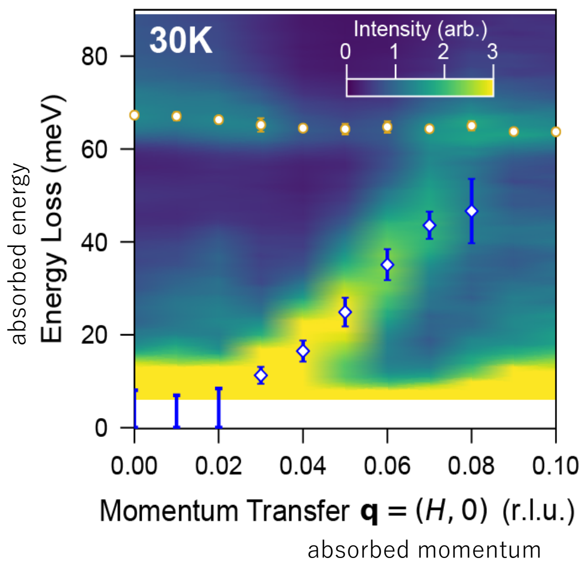

Figure 2. Newly observed electron excitation mode with zero energy gap. The 67 milli-electron volt signal is the optical phonon excitation of lattice vibrations. A typical plasmon excitation is observed at 1.2 electron volts (1,200 millielectron volts).

This achievement can be described as an accidental discovery: using a special experimental technique called M-EELS, the behaviour of electrons, including plasmons formed in metals, was directly observed, not by optical properties, but by striking electrons into the metal and measuring the momentum and energy of the reflected electrons. However, when the data was analysed, it contained an unusual excitation, the massless electron mode.

Figure 3. (Left) A single crystal of strontium-ruthenium oxide being grown by the floating zone method. http://trikuc.jp/research-topics/ (Upper) Grown single-crystal oxide metal.

Dr. Husain, now a research scientist at Quantinuum, USA, recalled, "At first, we had no idea what it was. Demons are not in the mainstream. The possibility came up early on, and we basically laughed it off. But, as we started ruling things out, we started to suspect that we had indeed found the demon."

The research group was joined by the theoretical group of their colleague Professor Philip Phillips from the University of Illinois, and Professor Phillips and postdoctoral researcher Edwin Huang performed calculations based on features of the electronic structure of strontium ruthenium oxides. They have shown that the observed massless mode indeed has the characteristics of a demon.

"Pines' prediction of demons necessitates rather specific conditions, and it was not clear to anyone whether strontium ruthenate should have a demon at all," Huang said. "We had to perform a microscopic calculation to clarify what was going on. When we did this, we found a particle consisting of two electron bands oscillating out-of-phase with nearly equal magnitude, just like Pines described."

Strontium ruthenium oxide is a material whose superconductivity was discovered by Professor Maeno and his colleagues some 30 years ago, but its superconductivity remains a mystery and has not yet been fully elucidated. By using previously little applied experimental methods with the aim of gaining a deeper understanding of the normal-conducting metallic state of this material, they have ventured into unexplored territory, resulting in an unexpected and significant discovery. According to Professor Abbamonte, "The results illustrate the importance of combining new methods with attractive materials and simply making unexplored measurements with precision. Most major discoveries are not planned. It's important to go to new places and see what's there."

3. Implications and future plans

As a future study, they will first investigate the Pines demon properties in strontium-ruthenium oxide. Experiments in which the directions of the momentum of the incident and reflected electrons were changed in various ways. It is necessary to examine and understand in detail how the nature of the demon changes.

In addition, there are likely to be many other materials in which Pines' demon mode can be observed. This electronic state, thought so special so far and overlooked, may actually be relatively common. Since this mode can be thermally excited, we will clarify what properties are reflected in some types of multiband metals. It is expected to have a ripple effect on the research that will be conducted.

4 About the research project

This research project was supported by the following funds:

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<Supplementary figure>

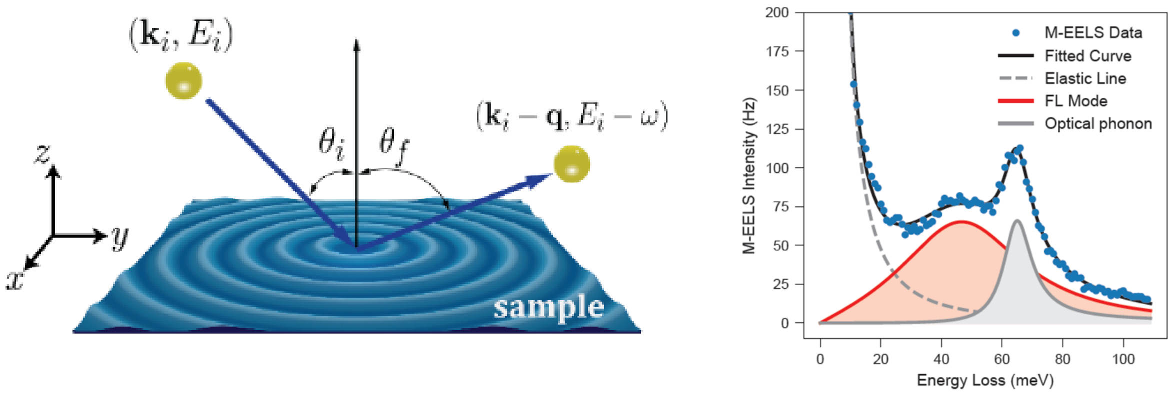

Figure 4. (Left) Conceptual diagram of incident and reflected electrons exciting a sample in momentum-resolved electron energy loss spectroscopy (M-EELS). (Right) An example of an observed demon mode spectrum. Gray peak is lattice vibration signal.