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Forefront in the Elucidation of the Mechanism of High-Temperature Superconductivity*

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The mechanism underlying the high-temperature (T_c) superconductivity of copper oxides has remained unelucidated and is one of the most difficult challenges of physics remaining in the 21st century. Various types of advanced spectroscopy have been employed to clarify the mechanism, resulting in the advancement of these techniques. Although the mechanism has not yet been completely clarified, the pseudogap phase, which always accompanies a superconducting phase, is now being considered as an electron state that plays a key role in the clarification of superconductivity.

1. Introduction

The mechanism underlying the high-temperature (T_c) superconductivity of copper oxides has remained unelucidated and is still one of the greatest mysteries in the field of condensed matter physics. A T_c of 135 K was observed in 1993 (164 K under high pressure); however, T_c has stopped rising since then. Although efforts to obtain higher T_c values appear to be faltering, many researchers have performed various types of experiments on material and physical properties and proposed theoretical models for the purpose of elucidating the mechanism underlying superconductivity.

Unprecedented amount of effort has been devoted to research on high-T_c superconductors in the last 24 years. Not only have a huge number of researchers been involved in this field but also experimental techniques have been diversified and experimental accuracy has markedly been improved, the trends of which continue to the present day. Currently, three types of spectroscopy are proving to be particularly useful in cutting-edge research on high-T_c superconductivity. In 1986, the highest energy resolution of photoelectron spectroscopy, one of the three types of spectroscopy, was only 100 meV. Today, it is 1 meV or better, and detailed electronic information on energy and momentum spaces can be obtained by angle-resolved photoemission spectroscopy (ARPES). Scanning tunneling microscopy/scanning tunneling spectroscopy (STM/STS) was originally used as a real-space probe with angstrom (Å) resolution; now it becomes possible to observe spatial patterns [spatial modulations of electron (hole) density] created by quantum-mechanical interference of de Broglie waves of electrons or superconducting quasi-particles. In neutron scattering spectroscopy, neutrons are used as a probe for the study of magnetism, which is considered to be closely involved in high-T_c superconductivity, and advances have been made in this technique. Not only electron-spin magnetism but also weak magnetism produced by the orbital motion of electrons can be captured by using spin-polarized neutrons.

2. Challenge of Investigating the Mechanism of Cooper Pair Formation

Clarification of the mechanism underlying high-T_c superconductivity is difficult because the number of electrons, as large as $10^{22}/cm^3$, exist with strong mutual repulsion in copper oxide. This strong mutual repulsion (referred to as strong electron correlation) prevents the motion of electrons. Therefore, electrons in the CuO_2 plane shown in Fig. 1 cannot move, which makes the CuO_2 plane a Mott insulator. At the same time, when the repulsion between electrons is strong enough, each electron becomes localized, and the localized spin magnetic moments form an antiferromagnetic order. The removal of electrons from the CuO_2 plane, i.e., doping of holes into the CuO_2 plane is necessary to make them mobile to increase the kinetic energy of electrons. However, in general, the motion of the doped holes is constrained by the antiferromagnetic order. Under such circumstances, it is very difficult to build "paths" where electrons/holes can freely move, and moreover develop them to "expressways", i.e., superconductivity.

Cooper pairs play an essential role in high-T_c superconductivity. The superconducting properties of high-T_c superconductors, such as zero resistance, the Meissner effect, flux quantization, and the Josephson effect, are the same as those of conventional superconductors. The difference between the two is in the d-wave symmetries of Cooper pairs of high-T_c superconductors. One direction for exploring the mechanism of superconductivity conventionally attempted by researchers is to determine the excitation (referred to as "glue") that mediates an attractive force between electrons, assuming that Cooper pairs are formed by mutual attraction between electrons. In conventional superconductors, lattice vibrations (phonons) can serve as the glue. Electrons (holes) that move freely in the CuO_2 plane interfere with each other; hence, the movement of electrons is strongly affected by other electrons. When electrons move, not only are phonons excited but also other excitations are induced. One of the possible excitations is spin excitations (antiferromagnetic fluctuations) with a large energy scale of about 0.15 eV. These excitations can be observed by several spectroscopy techniques. In copper oxides, phonons have equally high energy; therefore, it is difficult to distinguish between the excitation of phonons and spins. Furthermore, we have not found reasonable and acceptable answers to the basic questions of why the T_c value strongly depends on the doping level with spin excitations or phonons serve as a glue for the Cooper pairs.

Opposite to the first approach, in which it is assumed that Cooper pairs are formed by mutual attraction, an alternative approach for exploring the mechanism is to demonstrate that superconductivity can be brought about merely by repulsion without glue. This idea originated from the resonating

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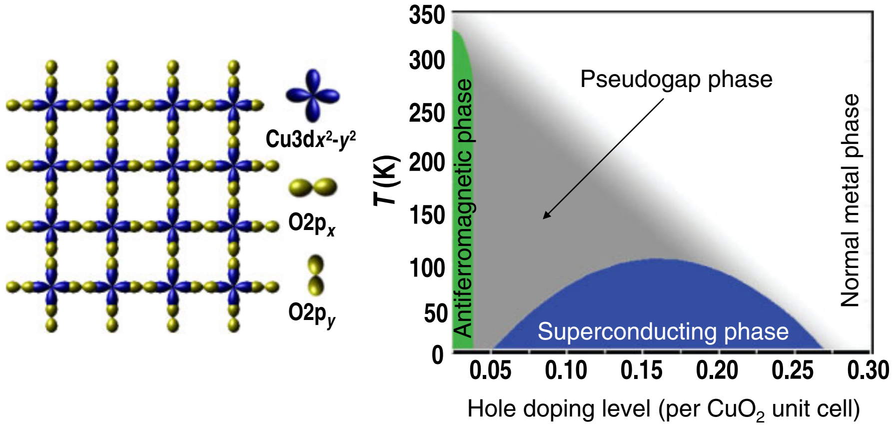

Fig. 1. (Color online) Two-dimensional square lattice (CuO_2 plane) consisting of copper (Cu) and oxygen (O) atoms (left) and evolution of electron phase with hole doping.

valence bond (RVB) theory proposed by P. W. Anderson. According to this idea, it is considered that the strong antiferromagnetic interaction between electron spins promotes Cooper pair formation. Whereas the first approach starts from the metal state, i.e., the overdoped region in the phase diagram of high-T_c superconductors, the second approach assumes that superconductivity is one of the states realized in a doped Mott insulator. Therefore, superconductivity strongly depends on the doping level. However, the type of experiment that can demonstrate the validity of this assumption has not yet been designed. I believe that it can only be validated by accumulating the results of a huge number of diverse experiments as circumstantial evidence,[1] which is also applied to the first approach.

Forefront researchers do not consider that clarification of the mechanism underlying Cooper pair formation will lead to clarification of the mechanism underlying high-T_c superconductivity. This is because the characteristics of superconductivity, i.e., 1) d-wave Cooper pairs, 2) low superfluid density (Cooper pair density), and 3) a strong two-dimensionality[2] can serve as factors to destabilize the superconducting state. Another reason is that the electron state and high-T_c superconducting state of the CuO_2 plane are far more complicated than widely anticipated; an unknown phase that is apparently competing with the superconducting phase has been found to coexist.

3. Clarification of Pseudogap Phase

3.1 Pseudogap

The previously mentioned unknown phase is generally called the pseudogap phase because energy gaps are observed in various spectra, such as those obtained by photoelectron spectroscopy and neutron scattering. Similarly to superconducting gaps and charge density wave (CDW) gaps, pseudogaps can provide the energy required to support the formation of long-range order over the entire crystal. The term "pseudo" has two meanings: 1) order is formed only locally (short range) instead of over a long range and 2) the relevant state is basically metallic although energy gaps are observed — an incomplete Fermi surface, called a Fermi arc, is observed.[3] The pseudogap state originally referred to the unusual "normal conducting state" that is realized between T_c and the pseudogap temperature, T^*, in the low-doping (underdoped) region. Recently, the coexistence of the pseudogap phase with a superconducting phase has been confirmed. The term "pseudogap phase" also refers to the region of dilute doping called the spin glass phase and the long-range stripe order of La-based superconductors. The stripe phase has been recognized as a different form of pseudogap owing to the unique properties of La-based superconductors.

The focus of recent research has been to clarify the pseudogap phase. In the phase diagram of high-T_c superconductors, the pseudogap phase occupies most of the area and even contains the superconducting phase. The pseudogap phase manifests itself 1) when the superconducting phase is not formed owing to too low doping, 2) when the superconducting order is destroyed at T_c by increasing the temperature, 3) when the superconducting order is weakened in the vicinity of the vortex core upon the application of a magnetic field, or 4) at the innermost CuO_2 plane of multilayer copper oxides with more than three CuO_2 planes in a unit cell. Many researchers agree that the mechanism underlying high-T_c superconductivity cannot be clarified without understanding of the origin and nature of the pseudogap phase and the relationship with the superconducting phase.

For the last several years, the electronic structure of the pseudogap phase and how the pseudogap phase coexists with the superconducting phase have been clarified through detailed experiments by STM/STS, ARPES, and neutron scattering.

(1) Two energy gaps with different magnitudes can be observed in the spectra of a material in the superconducting state obtained by various methods. The gaps with larger and smaller energies are considered to be a pseudogap (Δ_1) and a superconducting gap (Δ_0), respectively.[4]

(2) The electronic inhomogeneity of the superconducting state at the nanometer scale was reported as a result of an STM/STS experiment, which suggested that the phase of the CuO_2 plane may undergo separation into superconducting and pseudogap phases microscopically.[5] Since this experiment was carried out, new analytical techniques have been developed, and it has been revealed using this technique that

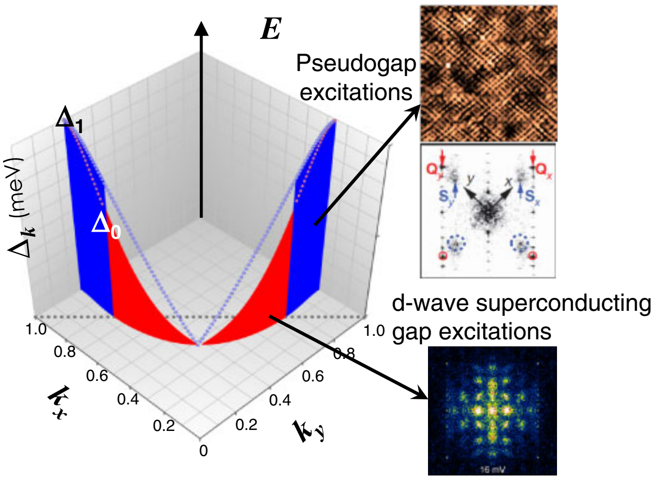

Fig. 2. (Color online) In the superconducting state, d-wave superconducting gap and pseudogap excitations are separated in momentum space. The red areas on the Fermi surface (line) with $E = 0$ represent the superconducting gap region. The superconducting quasi-particles excited across the superconducting gap (Δ_0) form quantum mechanical interference pattern (the Fourier-transformed spectrum is shown in the lower right figure). The blue areas represent the pseudogap region: the pseudogap has a larger energy (Δ_1) than the superconducting gap. The gap excitation produces a spatial density modulation pattern as shown in the upper right figure. The Fourier-transformed spectrum of the pattern is represented by dots Q (corresponding to Bragg reflection from a CuO_2 unit cell) and S (corresponding to stripe-like modulation).

the superconducting and pseudogap phases almost uniformly coexist on the CuO_2 plane.[6]

(3) Both phases behave differently in momentum space, although they homogeneously coexist on the CuO_2 plane. As shown in Fig. 2, a superconducting gap is observed in the area containing the center of the Fermi surface (red), whereas a pseudogap is observed in the outside areas (blue). When the temperature is increased or the doping level is lowered, the area of the superconducting region decreases and that of the pseudogap region increases. The former is observed by STM/STS as a quasi-particle interference pattern, and the latter is observed as a complicated pattern of electron density.[7] The high-T_c superconducting state is considered to be an unprecedented state where the pseudogap and superconducting phases homogeneously coexist in real space and undergo phase separation in momentum space.

3.2 Symmetry breaking

Recently, symmetry breaking in the pseudogap state has been discovered by various experimental probes. Spin excitation by neutron scattering[8] and electron transport phenomena[9] indicate anisotropy in the CuO_2 plane with a tetragonal structure having fourfold symmetry. It appears that the electron system spontaneously breaks the rotational symmetry to form an electronic structure with two-fold symmetry on the lattice with fourfold symmetry. A stripe pattern of an electron density modulation, the period of which is four times the lattice constant, can be observed in an STM/STS image of a $Bi_2Sr_2CaCu_2O_{8+\delta}$ crystal with a size of 1–2 nm.[7] In fact, the alignment of the stripe pattern is not uniform owing to the crystal disordering of Bi-based superconductors (Fig. 3). Some researchers refer to these broken symmetry states as electronic liquid crystals because an electronic liquid state has a preferential crystal orientation.

In a spin-polarized neutron scattering experiment, the presence of a weak antiferromagnetic order that breaks the time-reversal symmetry in the pseudogap state was reported.[10] Its magnetic structure is induced not by the electron spins but by the current due to the orbital motion of electrons of Cu and O atoms in the unit cell (Fig. 3).[11]

The breaking of spatial symmetry or time-reversal symmetry strongly indicates that the pseudogap state itself is a phase that conforms to a certain order different from that of superconductivity, rather than a precursor of the superconducting order.

By increasing the spatial resolution of STM/STS, the relationship between the electron density pattern and the location of atoms on a CuO_2 plane in the pseudogap phase was observed, which revealed the cause of symmetry breaking.[12] The reason for the lowering symmetry from fourfold to twofold is that O_x and O_y, the two oxygen atoms in a CuO_2 unit cell, which were originally equivalent, become electronically inequivalent. The type of inequivalence has not yet been clarified. However, in the antiferromagnetic order model using the current generated by the electron orbit, which is believed to induce the breaking of time-reversal symmetry, the two oxygen atoms in the CuO_2 unit cell are inequivalent. This fact could help clarify the microscopic mechanism underlying the symmetry breaking.

We have obtained reasonably satisfactory answers to basic questions on the pseudogap, i.e., whether a pseudogap state can be called a phase and how the pseudogap phase coexists and competes with the d-wave superconducting phase. Let us review the progress in the understanding on the pseudogap phase in terms of the mechanism underlying high-T_c superconductivity, i.e., whether the pseudogap phase is necessary for high-T_c superconductivity and how the pseudogap phase contributes to the realization of high-T_c superconductivity.

When the stripe phase is stabilized in La-based superconductors (Fig. 3), T_c significantly decreases. This indicates that there is a trade-off between the existence of the stripe order and that of the superconducting order. However, it has been found that the superconducting order develops from a high temperature in the CuO_2 plane even in a stripe phase.[13,14] With these factors, it is considered that the presence of a stripe phase does not inhibit electron pair formation within the plane; however, it inhibits the alignment of the phase of electron-pair wavefunction between neighboring planes, leading to a decrease in T_c.

Similarly, a phenomenon that can be explained by assuming the development of an in-plane superconducting order in the pseudogap state at temperatures higher than T_c has been discovered.[15] In fact, in an experiment on underdoped Bi2212 ($T_c = 37$ K) by STM/STS, superconducting quasi-particle interference in the CuO_2 plane was observed up to $T = 55$ K.[16] Note that the region of the superconducting phase decreases in the momentum space, compared with that of the material under the superconducting state at a temperature of T_c or lower. The pseudogap phase does not inhibit electron pair formation; however, it inhibits the alignment of the phases of pairs in the entire sample.

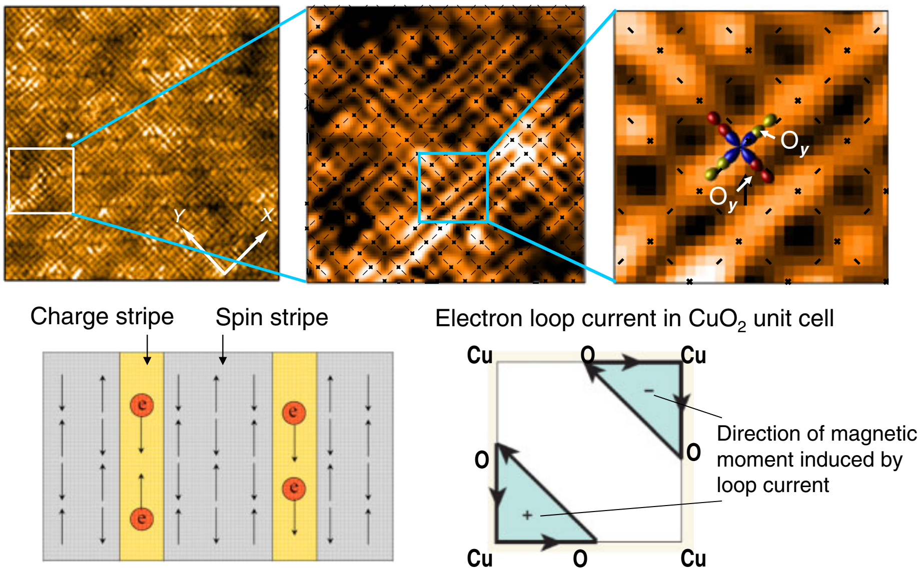

Fig. 3. (Color online) Upper figures: Spatial modulation pattern of electron density (upper left) induced by excitation in pseudogap phase. The electron density is high in the areas with lighter color. With increasing spatial resolution (upper middle and upper right), the electron density in the region between neighboring Cu atoms (indicated by cross marks) in the X-direction is found to be different from that in the Y-directions. In this region, there is an oxygen atom (O_x or O_y), and O_x and O_y are electronically inequivalent. Lower figures: Schematic drawing of stripe order (left) and loop-current order of electrons (right).

This finding suggests that the superfluid density ρ_s, rather than the superconducting gap, may be a factor that governs T_c in high-T_c superconductors, in contrast to that in low-T_c superconductors. ρ_s of copper oxides is at least one order of magnitude smaller than for low-T_c superconductors. ρ_s is also a measure of the phase rigidity of superconducting phase (originating from the quantum mechanical uncertainty relationship between the number of particles and their phases, $\Delta N \cdot \Delta\theta \sim 1$). Therefore, in superconductors with a low ρ_s, the phase rigidity inevitably decreases (or the phase fluctuation becomes large), leading to a decrease in the T_c value, at which the phase aligns.

In another paper, it was reported that the presence of the pseudogap phase strengthens the electron pairing interaction. The results of observing the $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ trilayer ($T_c = 110$ K) by ARPES revealed that the outer two CuO_2 planes are overdoped with holes while the inner CuO_2 plane is underdoped and has a very low hole concentration.[17] In monolayer and double-layer copper oxide superconductors, the overdoped plane should be in a superconducting phase with a small superconducting gap, and the underdoped plane be in a state in which the pseudogap phase dominates. However, experimental results indicate that the superconducting gap in the outer plane is much larger than expected and that the superconducting gap region in the inner plane greatly expands in the momentum space. In the multilayer superconductors, superconducting and pseudogap phases are thus in close contact to each other and become strongly coupled, which is assumed to be an important factor in the realization of superconductivity at T_c exceeding 100 K.

3.3 Remaining challenges

There is little doubt that the pseudogap plays a key role in the realization of high-T_c superconductivity. It will take more time before we can answer the following questions: which of the observed symmetry breakings can explain the mechanism of superconductivity, how symmetry breaking is related to electron pair formation, and why the symmetry breakings promote electron pair formation. It is currently considered that the mechanism underlying the high-T_c superconductivity of copper oxides may be clarified through a greater understanding of the pseudogap phase, a mystery in the field of high-T_c superconductivity.

Also, clarifying the mechanism underlying the overdoped region in the phase diagram is not an easy task, although this has not been discussed in this article. The results of several experiments indicate that the superconducting phase does not exist alone in the overdoped region; instead, it coexists with a normal metal phase (a Fermi liquid).[18] When multiple phases (orders) coexist, there is a possibility that strong fluctuations of a composite of spins, charges, and the orbitals of electrons, and phonons occur in the vicinity of the phase boundaries. The phase boundary which is theoretically proposed so far is the one between the pseudogap phase in the underdoped region and a normal metal phase in the overdoped region (which is called the quantum critical point[19] and is considered to exist at a doping level of $x = 0.16$–0.20, at which T_c is maximized).

As discussed earlier, the strong mutual repulsion between electrons prevents their movement. Assuming that the pseudogap phase refers to paths on which carriers doped into a Mott insulator can move, a superconducting phase can be considered as "expressways" on which electrons/holes are paired so that carriers can move more freely. The superconducting phase realized by d-wave Cooper pairs at a low superfluid density is extremely fragile; one option to stabilize the superconductivity is to adopt the pseudogap phase as a "bypass".

4. Conclusions: Impact on Applied Physics

The research on high-T_c superconductors has encouraged the remarkable development of the following two factors; 1: the spectroscopy techniques as a means of evaluating material properties, such as ARPES and STM/STS, 2: an imaging technique to visualize clearly the huge amount of detailed data on the electron structure and electron excitation obtained through these techniques. These techniques are expected to be applied to various fields in applied physics and to manifest their power by contributing to developments in these fields.

The most significant impact of the high-T_c superconductors on applied physics is that superconductivity has been realized at temperatures higher than the boiling point of liquid nitrogen; even the existence of a room-temperature superconductor has been discussed seriously rather than as a dream. Although they have scarcely been considered in the case of low-T_c superconductors, the properties of high-T_c superconductors such as high anisotropy, the unique symmetry of the Cooper pairs, and low superfluid density, as discussed in this review, have attracted interest from researchers. The development of a new concept for superconductors will be necessary when considering the mechanism underlying high-T_c superconductivity and its applications.

If room-temperature superconductivity were realized in copper-oxide- or iron-based superconductors, it would be an important issue to discuss about what is the constraint on the practical application. Researchers agree that s-wave pairing is more suitable for device application because the inelastic scattering of electrons becomes more active with increasing temperature. From the viewpoint of critical current (J_c), anisotropy and low superfluid density will be problems. For example, with increasing anisotropy, J_c in high-T_c superconductors is expected to sharply decrease. One of the future challenges in applied physics will be to discover isotropic room-temperature superconductors with s-wave Cooper pairs. Superconductors with weak anisotropy can be used at the temperature of liquid nitrogen.

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