様式３

論　文　内　容　の　要　旨

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論文題名	Pseudogap and precursor superconductivity study of $YBa_2(Cu_{1-x}Zn_x)_3O_y$: *c*-axis optical study from underdoped to overdoped region （銅酸化物高温超伝導体$YBa_2(Cu_{1-x}Zn_x)_3O_y$の擬ギャップと超伝導の前駆現象の研究：不足ドープ領域から過剰ドープ領域におけるc軸光学スペクトル）

論文内容の要旨

In this study, the charge dynamics of the high temperature cuprate superconductor $YBa_2(Cu_{1-x}Zn_x)_3O_y$ over the electronic phase diagram have been investigated in a wide doping range, as well as, for several Zn-contents, both in the normal and the superconducting state. The temperature dependent reflectivity measurements have been performed with Fourier transform infrared spectroscopy and the optical conductivity spectra have been calculated from the reflectivity by using the Kramers-Kronig transformation.

Detailed spectral weight analysis was performed on the optical conductivity spectra, especially in the pseudogap region. Several issues regarding to the electronic phase diagram were discussed. As a result, we obtained several important conclusions. Firstly, the behavior of the pseudogap and the relation between the pseudogap and the superconducting gap, secondly, the precursor of superconductivity, and finally, some remarks related to the kinetic energy driven superconductivity were discussed.

Pseudogap:

Pseudogap has a very clear behavior in the optical conductivity spectra. With pseudogap opening, the low energy optical conductivity is suppressed and the lost spectral weight (SW) is transferred to the high-energy region. With superconducting gap opening similar low energy response can be seen, however, the lost spectral weight is redistributed at delta-function at zero frequency. Hence the pseudogap behavior and the superconducting behavior can be resolved within a single measurement in a *c*-axis polarized optical conductivity.

We trace the SW as the pseudogap response in the superconducting region. The analysis showed that the SW that is transferred to the high-energy region with pseudogap opening does not contribute to the superconducting condensation. This gives evidence to the fact that the pseudogap is not the precursor of the superconductivity. Moreover, the high-energy transfer of the SW, which is the indication of the pseudogap behavior, continues even below the superconducting transition temperature (T_c) indicating the coexistence of the pseudogap and the superconducting gap in the superconducting state.

With the evidences given above, the Zn-effect on pseudogap and the pseudogap behavior in the overdoped region of the phase diagram give further evidence to the different order and coexistence scenario. We observed that the pseudogap is robust to Zn-substitution and even can be observed for a non-superconducting sample (the sample that we suppressed superconductivity with Zn-substitution). Moreover, in the overdoped region, pseudogap temperature (T^*) crosses the superconducting dome and goes to zero around 20% carrier doping, while superconductivity survives up to higher doping levels.

As a result, the SW analysis, Zn-doping effects, and the behavior in the overdoped region leads to the conclusion that the pseudogap is not the precursor of the superconductivity and should be thought as another order. Moreover, the pseudogap and the superconducting gap coexist in the superconducting state.

Precursor superconductivity:

Even though we eliminated the pseudogap as the precursor of the superconductivity, another temperature scale can be defined as the precursor state. Previously many experimental probes observe this state, as well. However, there is no consensus about the temperature range and the doping dependence of the precursor superconductivity. One group of experiments showed that the precursor state extends to high temperatures (3-4 T_c) with a rather unusual doping dependence. Other group of experiments, on the other hand, presented that the precursor state extends up to close vicinity of T_c and closely follows the doping dependence of the T_c.

In this study, temperature dependent superfluid density is investigated from the real and the imaginary part of the optical conductivity. The calculations showed the existence of the superfluid density starting from the temperatures higher than T_c (We marked as T_p). With decreasing temperature, superfluid density gradually increases and at a temperature T_c' near T_c, the slope of the increase suddenly becomes steeper. It seems that the superconducting carriers persist up to much higher temperatures than T_c, although its fraction is very small (less than a few % of the total superconducting carrier density at T =0).

It is interesting that the doping dependences of T_c' and T_p are different. T_c' is always 10-20 K above T_c and thus follow the T_c change, whereas T_p increases with decreasing doping levels (p) and reaches much higher temperatures than T_c'. T_c' and T_p are almost merged at the optimum doping p = 0.16. On the other hand, with Zn-substitution both temperature scales decrease like T_c, unlike T*. The Zn-dependence confirms that these features are related with superconductivity. The observation of the superfluid density suggests that the superconducting regions start to appear at T_p. With decreasing temperature these regions expand and start to see each other leads to the system to the bulk superconductivity starting at T_c', which can be explained in the framework of the inhomogeneous superconductivity.

Kinetic energy driven superconductivity:

There are some theories that explain the superconductivity mechanism as the kinetic energy driven superconductivity. Previously, while the c-axis optical conductivity was studied, it was observed that the superfluid density obtained by the missing area method is significantly smaller than that calculated from the imaginary optical conductivity. It has been explained that some of the carriers contributing to the condensation actually come from very high-energy region (around visible region), which corresponds to the kinetic energy contribution. However, in many cases, the missing area is underestimated, since it has been calculated within the limited energy region. The calculated condensation energy for the *c*-axis spectra is smaller than the theoretical expectations. Therefore, in the early days it has been thought that the remaining portion might be observed in the in-plane spectra. It is necessary to point out that this is only observed for the Bi2212 system in the in-plane spectrum, and other systems do not show such kind of high energy contribution.

We have revealed that the discrepancy of the superfluid density does not come from the kinetic energy contribution, but it is due to the mistreatment of the so-called transverse Josephson plasma (TJP) mode. We observed that in the case of suppression of the TJP mode with Zn-substitution, the superfluid density discrepancy disappears. The TJP mode creates a broad absorption peak in the spectrum, which causes the underestimation of the missing area and leads to the discrepancy of superfluid density.

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論文審査の結果の要旨及び担当者

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論文審査の結果の要旨

本論文は、銅酸化物高温超伝導体の電子状態解明において、最大の課題となっている「擬ギャップ現象」について、Zn 置換した $YBa_2Cu_3O_{7-\delta}$の c 軸偏光光学スペクトルを用いて研究した結果をまとめたものである。

2.5meV から 20eV までの広いエネルギー範囲に渡る光学反射スペクトルの詳細な温度依存性、組成依存性を精密に測定した結果、(i)擬ギャップ形成に関与したキャリアは、超伝導凝縮しないこと、(ii)超伝導転移温度以下でも擬ギャップが開き続けること、(iii)Zn 置換により超伝導性が消失した試料においても擬ギャップが観測されること、(iv)過剰キャリアドープ領域では超伝導転移温度以下に擬ギャップ温度が存在すること、等がわかった。擬ギャップの起源については、これまで①超伝導前駆現象であるとする説と②超伝導とは競合する別の秩序状態であるとする説、の間で論争があった。本研究の(i)-(iv)の結果はすべて、前者を完全に否定し、後者を支持するものである。

また近年、擬ギャップ温度より低温の常伝導状態において、超伝導前駆現象が観測されるという報告もいくつかなされている。しかし、測定プローブによって、前駆現象が観測される温度やその組成依存性が異なっており、実験結果の統一的理解にはほど遠い状態である。本研究では、得られた光学伝導度スペクトルの強度の温度依存性から、超伝導転移温度よりかなり高温から超伝導凝縮成分が存在していることが見出された。超伝導凝縮成分（超伝導キャリア）を光学伝導度の虚部から見積もっても、同じ温度から有限の値が観測された。この特徴的温度 T_p は、Zn 置換により超伝導転移温度が低下すると、それにスケールして低下し、この現象が超伝導前駆現象であることを強く支持した。

キャリアドープ量が減少すると超伝導転移温度は低下するが、T_p は逆に上昇することがわかり、本物質の超伝導対形成機構が、モット絶縁体をもたらす強い電子相関に起因していることを強く示唆する結果となった。これまで前駆現象として報告されていた温度の一部は、通常の超伝導揺らぎ理論で理解できるものであることも示され、過去の混沌とした実験結果を統一的に整理して理解することに成功した。

以上のように、本研究は物性物理学の長年の課題である銅酸化物高温超伝導体の電子状態解明、超伝導機構の解明に大きく貢献するものである。丁寧な試料作製、精密な光学測定、詳細なデータ解析によって導き出された結果は、独創性に優れ、当該分野の研究として高く評価できる。

よって本論文は、博士（理学）の学位論文として十分価値のあるものと認める。