Spatial inhomogeneity in $Bi_2Sr_2CaCu_2O_{8+\delta}$ investigated by micro photoemission spectroscopy

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In recent years strongly correlated electron systems have attracted a great deal of attention because they exhibit interesting phenomena such as high- T_c superconductivity and giant magnetoresistive effect. These properties are caused by the competition and/or coexistence of multiple interactions (many-body interactions), such as those between electrons and bosons (phonons, magnons, etc.), due to strong electron correlation. In addition, it has recently become clear that the physical properties of such complex electron systems are based on "the self-organization of electrons", in which the electron system is ordered on a specific spatial scale and size of relevant interaction [1]. On the other hand, the theoretical treatment of many-body interactions is always an enormous challenge. It is otherwise approximated with the mean field and sometimes leads to wrong interpretations. Therefore, it is necessary to investigate electronic states and their many-body interactions experimentally. Photoelectron spectroscopy is a powerful experimental technique that can satisfy such requirements. In the past, the spatial resolution of photoemission spectroscopy was as low as about a millimeter, but in recent years, micro photoemission spectroscopy with well-improved spatial resolution has been developed worldwide [2,3]. In fact, in Cr-V₂O₃, a prototypical strongly correlated electron system, the inhomogeneous distribution of metallic and insulating electronic phases on a scale of several tens of microns has been observed by micro photoemission spectroscopy [4]. Such electronic phase separation is an essential phenomenon of strongly correlated electron systems, which is not found in conventional metals, insulators, and semiconductors. Therefore, it is essential to observe the electronic state with high spatial resolution in order to understand the physical properties of strongly correlated electron systems.

In this study, we have developed a micro angle-resolved photoemission spectroscopy (micro-ARPES) system at the Hiroshima Synchrotron Radiation Center, Hiroshima University, and investigated the real-space behavior of the electronic state of the cuprate high- T_c superconductor Bi₂Sr₂CaCu₂O_{8+ δ} (Bi2212). In addition to the valence band information, we also carried out micro X-ray photoelectron spectroscopy (micro-XPS) measurements to analyze the chemical state. Also, we analyzed the spatial mapping data using a clustering method based on machine learning to handle big data, usually obtained in microscopy.

Figure 1(a) shows the band dispersion of Bi2212 measured by micro-ARPES. In this study, we evaluated the position dependence of the ARPES spectra by measuring the band dispersion at each position with small changes in the measurement position. Next, the momentum dependence of the band dispersion was measured at two points (A and B) where the sample positions were slightly different (0.2 mm), and the different sizes of the superconducting gap were observed [Fig. 1(b)]. This result suggests that the doping level differs depending on the position, and there is a small but finite difference in the observed size of the Fermi surface. On the other hand, it is still not realistic to examine the spatial dependence of such detailed and fine electronic structures by high-resolution ARPES because of time constraints. Therefore, we have performed micro-XPS measurements enabling microscopic measurements in a wider range with finer steps.

In this study, two-dimensional spatial mapping of the O 1s spectrum was performed by micro-XPS. The mapping range and step were respectively set to 1 mm and 5 μ m each in horizontal and vertical directions, providing 40,000 O1s spectra in total. Since the total measurement time had to be limited to suppress the spectral change due to the surface degradation, the statistical accuracy of the experimental data at each measurement point was insufficient, making peak analysis difficult [FIG. 2(a)]. In addition, it is difficult to understand the characteristics of the electronic state only from the two-dimensional spatial distribution of the integrated intensity of the O 1s spectrum, as shown in FIG 2(c). Therefore, we performed a clustering analysis using machine learning on the spatial mapping data. The two-dimensional spatial distribution of each cluster obtained by clustering is shown in FIG. 2(d). At first glance, it is clear that each cluster is distributed inhomogeneously near the center of the sample, unlike the spatial distribution of the integrated intensity. By integrating the whole data classified into the same cluster, the obtained spectrum has enough statistical accuracy for peak analysis, making it possible to extract the electronic features, as exemplarily shown in FIG. 2(b). The subsequently performed peak analysis on each integrated spectrum [FIG. 2(e)] revealed that the peak positions shifted by more than 0.1 eV between clusters. This result clearly indicates that the chemical potential shift depending on the position, demonstrating the presence of spatial inhomogeneity in the doping level.

The present micro-ARPES and micro-XPS studies consistently showed that cuprate high- T_c superconductor Bi2212 exhibits micro-scale electronic inhomogeneity. Our results provide an important guideline for understanding the phase transition phenomena in high- T_c superconductivity.



FIGURE 1. (a) ARPES image along the nodal direction (Γ Y direction: blue line in FIG. 1(d)). (b) Momentum dependence of the superconducting gap measured at two points A and B (see FIG. 1(c)). (c) Optical microscope image observed ex-situ after ARPES measurements. (d) A quarter Brillouin zone of Bi2212, where θ is the Fermi surface angle defined by two segments starting from the Y point (π , π). Here, $\theta = 0$ corresponds to the nodal direction.



FIGURE 2. (a) Raw O 1s spectrum at one measurement point acquired by two-dimensional spatial mapping. (b) O 1s spectrum obtained by integrating data classified into the same cluster by clustering analysis. (c) Two-dimensional spatial distribution of the integrated intensity of the O 1s spectrum. (d) Two-dimensional spatial distribution of clusters. (e) Integrated spectra of each cluster (squares) and results of peak analysis (lines). The circles represent the peak position determined by fitting.

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