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High-resolution photoemission study of low-T_c superconductors: Phonon-induced electronic structures in low-T_c superconductors and comparison with the results of high-T_c cuprates

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Abstract

Recent advances in the energy resolution and cryogenic techniques used in photoemission spectroscopy have enabled us to directly observe the superconducting electronic structure of superconductors with a relatively lower transition temperature (low-T_c). In this paper, we report photoemission results on low-T_c superconductors measured with an ultrahigh-resolution low-temperature photoemission spectrometer. The result of Nb_3Al exhibits a superconducting gap and a phonon-induced peak–dip–hump structure characteristic of the strong-coupling superconducting spectral function. From measurements of $Ba_{0.67}K_{0.33}BiO_3$, we observe isotropic s-wave superconducting gap opening below T_c as well as depletion of intensity near the Fermi level above T_c, which can be attributed to a phonon-induced pseudogap. We will compare these photoemission results with those of high-T_c superconductors.

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1. Introduction

It is generally accepted that phonon mediates superconductivity in conventional superconductors [1]. For high-T_c superconductors, exotic mechanisms other than of phonon-origin have been speculated [2], because the strong electron–electron correlation originating in Cu 3d electrons has been thought to play an important role for the superconductivity occurring at high temperatures. As for the electronic states of the cuprates, photoemission spectroscopy has been used

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to reveal the normal and superconducting electronic structures that give insight into the mechanism of superconductivity, e.g. the $d_{x^2-y^2}$-wave superconducting gap [3], and the small pseudogap [4], and the large pseudogap [5,6].

In contrast, photoemission study of conventional superconductors has been difficult till very recently due to the limitation of energy resolution and cryogenic techniques used in photoemission spectrometers, since the energy scales of conventional superconductors having lower T_cs is smaller (few meV) compared to those of high-T_c cuprates (several tens of meV). Consequently, lack of reference spectra of conventional superconductors has hindered simple comparisons that lead to clarify what are the distinctive electronic features of high-T_c cuprates. Therefore, it is essential to study the electronic structures of conventional low-T_c superconductors in both superconducting and normal states. From such a study, we would be able to know the features induced by phonons in conventional superconductors and can directly compare them with the high-T_c results in order to know the inherent electronic features of high-T_cs.

In this paper, we present high-resolution photoemission results of Nb_3Al and $Ba_{0.66}K_{0.33}BiO_3$ (BKBO). Superconducting electronic structure of Nb_3Al shows a peak–dip–hump structure characteristic of the strong-coupling spectral function. Temperature-dependent photoemission spectra near the Fermi level (E_F) of BKBO show clear s-wave gap opening across the superconducting transition. Moreover, those measured on a wider energy scale manifest the existence of a pseudogap with a lager energy scale (~70 meV) than that of the superconducting gap (~5 meV), which is filled up on increasing temperature. These results are compared with the results of high-T_c cuprates and some implications are discussed.

2. Experimental

Polycrystalline Nb_3Al samples were prepared by an arc-melting method. Single crystals of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) have been grown with a traveling solvent floating zone method [7]. Single crystals of $Ba_{1-x}K_xBiO_3$, with $x = 0.33$ were prepared by an electrochemical method and characterized as reported earlier [8]. Magnetization measurements exhibited sharp transitions with onset temperatures at 17.8 K for Nb_3Al, 91 K for Bi2212 (optimally doped), and 31 K for $Ba_{1-x}K_xBiO_3$.

Photoemission measurement were performed on a spectrometer built using a GAMMADATA-SCIENTA SES2002 electron analyzer, a high-flux discharging lamp with a toroidal grating monochromator, and a liquid-He-flow-type refrigerator with a newly designed thermal shield. The total energy resolution (analyzer and light) using He Iα (21.218 eV) resonance line was set to ~4 meV for superconducting-state measurements of Nb_3Al and BKBO, and set to ~8 meV for normal-state measurements of BKBO and superconducting-state ones of Bi2212. The sample temperatures were measured using a silicon-diode sensor mounted just close to a measured sample. The base pressure of the spectrometer was better than 5×10^{-11} Torr. Samples were fractured for Nb_3Al, cleaved for Bi2212, and scraped for BKBO in situ to obtain clean surfaces. For each sample, E_F of samples was referenced to that of a gold film evaporated onto the sample substrate.

3. Results and discussion

Nb_3Al is an A15-type superconductor with strong electron–phonon coupling [9]. In Fig. 1, a high-resolution photoemission spectrum of Nb_3Al measured at 5.5 K (superconducting state) is shown within a small intensity window in order to highlight structures appearing on a larger energy scale compared to the size of the superconducting gap. Besides the superconducting condensation peak at 5 meV, we clearly observe dip and hump structures around 25 and 40 meV, which are the typical features of the strong-coupling superconducting spectral function [1]. The observation of the peak–dip–hump structure is consistent with the reduced gap parameter $2\Delta(0)/k_BT_c$ of 4.0 estimated from the present study and also agrees well with previous transport and tunneling measurements, which classified Nb_3Al into a strong-coupling superconductor [9].

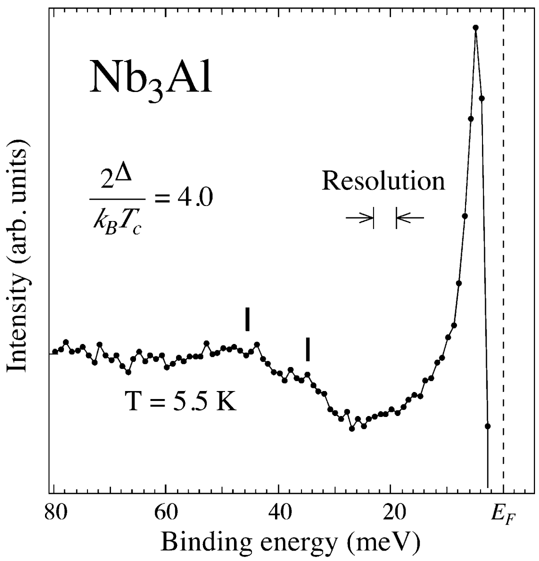

Fig. 1. High-resolution photoemission spectrum of Nb_3Al at 5.5 K shown in a limited intensity window.

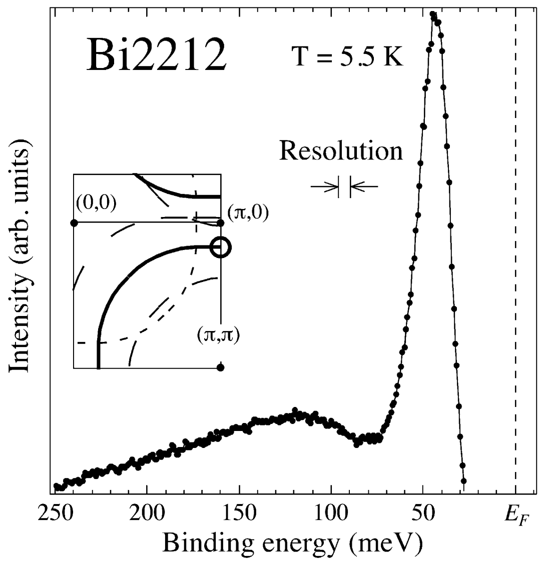

Fig. 2. High-resolution angle-resolved photoemission spectrum of optimally doped Bi2212 at 5.5 K measured at a E_F crossing point along the $(\pi, 0)$–(π, π) direction, which is shown in a limited intensity window.

The peak–dip–hump structure has been reported in the high-temperature superconductor Bi2212 near the $(\pi, 0)$ point in the Brillouin zone [10,11], and has been actively discussed in relation to the magnetic resonance mode observed by neutron measurements [11,12], and more recently in relation to a characteristic phonon mode [13]. Here we try to compare the results of Nb_3Al with that of Bi2212. In Fig. 2, we show angle-resolved photoemission (ARPE) spectrum obtained at Fermi momentum close to the $(\pi, 0)$ point, where the $d_{x^2-y^2}$-wave order parameter has the largest magnitude. The peak–dip–hump structure is seen with the peak and dip being located at 43 and 80 meV, respectively, consistent with previous reports [10,11]. The overall shape of Bi2212 looks similar to that of Nb_3Al regardless of the difference in energy scale. However, we also notice differences. First, the width of the peak of Bi2212 is very broad compared to the resolution we used, while that of Nb_3Al is very sharp and indeed resolution limited. Non-resolution-limited coherent peak width has been clarified by ARPE study using a resolution of 8 meV [14], where consistency with optical studies has been mentioned. According to the most recent scanning tunneling microscopy/spectroscopy [15], the superconducting gap shows a microscopic electronic inhomogeneity in Bi2212, while the gap of a conventional superconductor like Nb is spatially homogeneous. The broadened width of ARPE peak in Bi2212 and the resolution-limited peak in conventional superconductor Nb_3Al shows a close connection between the width and the spatial inhomogeneity. Secondly, the overall spectral shape beyond the peak of Bi2212 looks also rather broad compared to that of Nb_3Al, where fine structures originating in structures in $\alpha^2F(\omega)$ are clearly observable (note two weak structures at 35 and 45 meV in Fig. 1). This observation seems to imply that the spectral shape of Bi2212 is dominated by a coupling of electron with one strong mode, as has been speculated [10,11,13]. However, since there is tunneling results of Bi2212 showing fine structures that can be compared with phonon density of states [16], further ARPE study with extremely high signal-to-noise ratio is worth trying.

Next we move on to photoemission study of BKBO. BKBO is a perovskite three-dimensional superconductor having relatively higher T_c exceeding 30 K [17]. While the mother compound $BaBiO_3$ is an insulator with a charge-density-wave

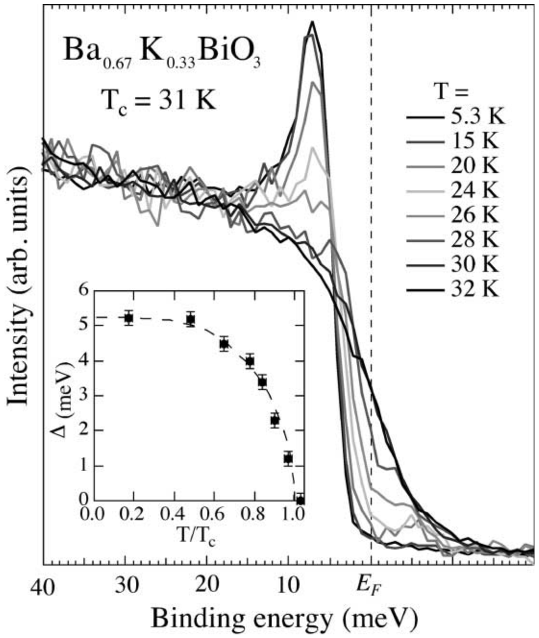

Fig. 3. Temperature-dependent high-resolution photoemission spectra near E_F of BKBO. Inset shows superconducting gap values as a function of temperature, as compared with a BCS prediction.

gap of ~2 eV [18], it becomes a superconductor on hole doping with a substitution of K for Ba, reminiscent of the phase diagram of high-T_c, though the origin of the insulating phases are different. In Fig. 3, we show temperature-dependent photoemission spectra of BKBO. We clearly observe redistribution of spectral weight from the region around E_F to higher binding energy, accompany by appearance of new peak around 7 meV, with decreasing temperature. These spectral modifications clearly demonstrate the opening of the superconducting gap below T_c. From the 5.3 K spectrum, we found that the leading edge is very steep, which strongly indicates that the superconducting order parameter of BKBO is isotopic s-wave, consistent with previous tunneling measurements [19]. The superconducting gap value determined from a Dynes analysis is 5.2 meV, resulting in the reduced gap value $2\Delta(0)/k_B T_c$ of 3.9, which classifies BKBO into a moderately strong-coupling superconductor. Further, we found that the normal-state spectrum of 32 K can be well reproduced by a Fermi–Dirac (FD) function of 32 K convolved with the known energy resolution, indicative of no pseudogap having the same energy scale as the superconducting gap. In Bi2212, there is a small pseudogap having a similar energy scale to and the same symmetry as the superconducting gap [4], which is most likely ascribed to pairing above T_c without phase coherence [20]. Observation of no small pseudogap in BKBO is consistent with the superconducting fluctuation models, where the fluctuation is expected to be stronger in two dimension than in three dimension [21].

More importantly, we measured temperature-dependent photoemission spectra on wider energy scales (Fig. 4) and found that there is a pseudogap above T_c. For 5.3 and 32 K spectra, we observe a suppression in the spectrum near E_F, though, at higher temperatures, the pseudogap feature is less clear due to thermal excitation of electrons. In order to remove the effect of the FD function, we divide the raw data by the corresponding FD functions (inset). Now we see that the pseudogap

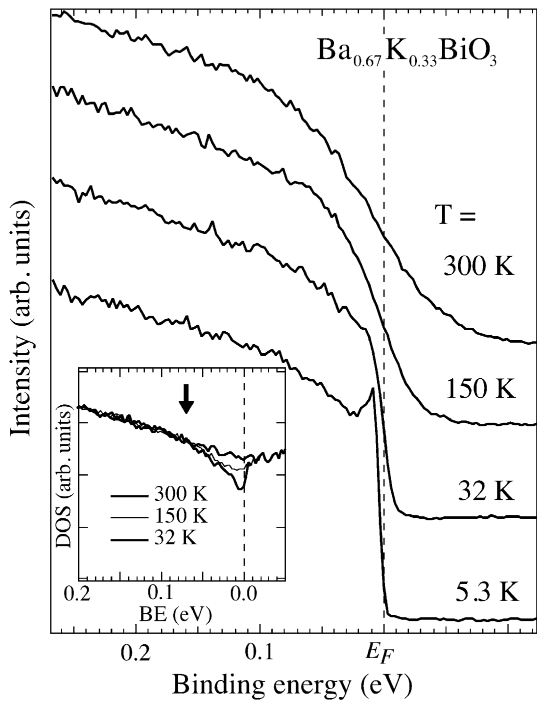

Fig. 4. Temperature-dependent high-resolution photoemission spectra measured over a wider energy range of BKBO. Inset shows temperature-dependent density of states as obtained from a division of raw data by a corresponding FD function.

has an energy of ~70 meV, which matches the energy of the breathing mode phonon [22] located around the highest phonon energy [23], and that it also fills in as temperature is increased. The observation of anomalous temperature-dependent pseudogap in BKBO suggests that the breathing mode phonon strongly coupled to the electrons in BKBO even in the metallic phase.

In high-T_c, photoemission measurements have revealed two pseudogaps in the normal state; one is the small pseudogap having the same energy scales of superconducting gap [4] and the other is a pseudogap having larger energy scales compared to that of the small pseudogap [5,6]. The large pseudogap showing temperature dependence has been observed from angle-integrated photoemission study of $La_{1.85}Sr_{0.15}CO_4$ [5] and Bi2212 [6]. The temperature where the large pseudogap fills in can be compared to the characteristic temperatures from magnetic measurements [24,25] and therefore speculated to be a magnetic origin. However, observation of similar temperature-dependent large pseudogap in a non-magnetic compound seems to request further consideration including a role of phonon to the origin of the larger pseudogap.

4. Summary

In summary, we have shown high-resolution photoemission results of Nb_3Al and BKBO. In superconducting-state spectrum of Nb_3Al, we observe a peak–dip–hump structure characteristic of the strong-coupling superconducting spectral function. Further, we found that the width of the peak of Nb_3Al is resolution limited, in sharp contrast to that of Bi2212, where the width of the coherent peak is very broad. For BKBO, we observe a pseudogap having the same energy scale of the breathing mode phonon and showing substantial temperature dependence, in very similar manner of the large pseudogap observed in high-T_c cuprates. These observations suggest that further photoemission study on phonon-mediated superconductors might clarify the phonon-induced features in conventional phonon-mediated superconductors and might lead to a deeper understanding of the mechanism of high-temperature superconductivity.

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