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Synchrotron Radiation PES Studies of High T_c Superconductor Surfaces and Interfaces for Contact and Junction Formation

Masaharu OSHIMA, Yasuko YAMADA, Tomoaki KAWAMURA, Satoshi MAEYAMA, Kouji HOHKAWA*, Yasuo TAZOH*, and Tsuneaki MIYAHARA**
NTT Applied Electronics Laboratories, Musashino, Tokyo 180, Japan
* NTT LSI Laboratories, Atsugi, Kanagawa 243-01, Japan
** Photon Factory, KEK, Tsukuba, Ibaraki 305, Japan

To fabricate devices such as Josephson Junction devices using recently developed high Tc superconducting materials, interface characteristics between contact metals or junction materials and superconductors were analyzed with synchrotron radiation photoemission spectroscopy. A new surface processing technique consisting of O^+ ion sputtering and in situ Au deposition processes on $YBa_2Cu_3O_Y$ was developed to realize very low contact resistance based on these SRPES experiments.

1. Introduction

To fabricate devices such as Josephson Junction devices using high Tc superconducting materials, interface characteristics between contact metals or junction materials and superconductors should be clearly understood. So far, there are only a few studies reported concerning superconductor interfaces. Hill et al[1] reported oxygen withdrawal and interface reaction for Fe/LaSrCuO. Furthermore, Gao et al[2] reported interfacce formation of high Tc superconductors with noble metals, transition metals and Ge. However, there are no papers reported concerning dry cleaning processes for device application.

In this study, surfaces and interfaces of high Tc superconductors were analyzed from the viewpoints of (1) development of a new dry process with maintaining surface superconductivity, (2) Ohmic contact formation, and (3) junction formation with synchrotron radiation photoemission spectroscopy which enables very surface sensitive analysis and resonance photoemission analysis.

2. Experimental

Experiments were carried out at BL-1A of the Photon Factory in KEK. The energy of photon monochromatized by GCM (Grating/Crystal Monochromator)[3,4] was from 54 to 125 eV. Samples were $YBa_2Cu_3O_Y$ bulk materials(Tc=90K) and $YbBa_2Cu_3O_Y$ thin films(Tc=76K) on MgO(100) substrates. Surfaces were cleaned by scraping with a special file in UHV(10^{-10} Torr). Dry cleaning processes such as Ar^+ and O^+ ion sputtering and oxygen plasma treatment by DC glow discharge were performed. Au and Si were deposited on scraped clean surfaces at room temperature at a deposition rate of about 0.3 Å/sec measured by a quartz thickness monitor.

3. Results and discussion

Valence band spectra of the $YBa_2Cu_3O_Y$ surafce cleaned by in vacuum file-scraping method were measured with different photon energy between 54 and 125 eV. When the incident photon energy was tuned to about 76 eV, a peak which appeared only on Cu^{2+} ($3d^9$

configuration) materials owing to resonance photoemission[5] came up at a B.E.(binding energy) of about 12 eV on the clean $YBa_2Cu_3O_Y$ surface, as shown in Fig.1. The peak which appeared at the same B.E. also on the XPS spectra is probably due to Cu MVV Auger transition. The mechanism of this Cu3p-3d resonance photoemission is shown in Fig.2. Thus this implies that the resonance peak can be used to check whether or not the very surface region (less than 10 Å) is in the Cu $^{2+}$ state. When the clean surface was Ar-sputtered, this peak disappeared completely and Cu^{2+} was reduced to the metallic state. This means that Ar ion bombardment causes Cu-O bonding to break, and oxygen to be removed.

On the other hand, O^+ ion sputtering was found to effectively clean the surface with maintaining the resonance peak. Figure 3 shows valence band spectra of $YbBa_2Cu_3O_Y$ thin films as-received and O^+ ion sputtered in comparison with the scraped clean $YBa_2Cu_3O_Y$ surface. The as-received surface is regarded as a little semiconductor-like state, because valence band maximum is deeper by about 0.9eV

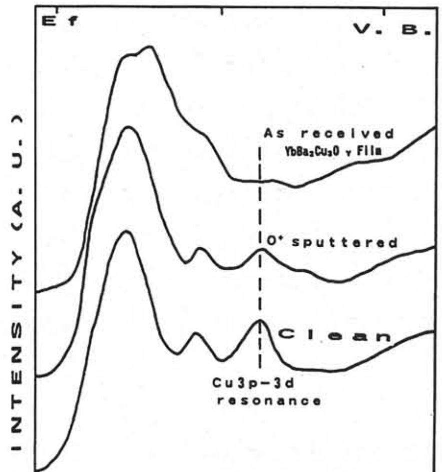

Fig.3 Valence band spectra of $YbBa_2Cu_3O_Y$ thin films as-received and sputtered by O^+ ions in comparison with scraped clean $YBa_2Cu_3O_Y$ surface. $h\nu$=76 eV.

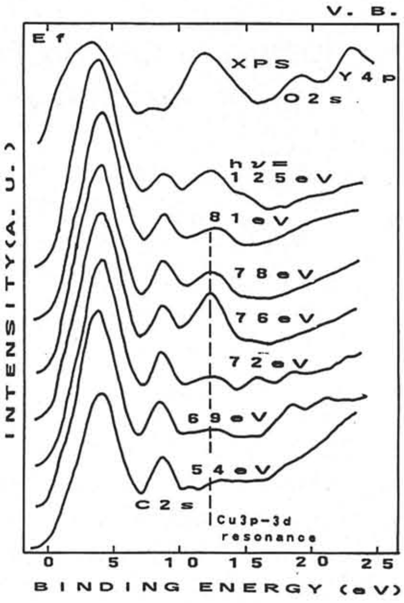

Fig.1 Valence band spectra of $YBa_2Cu_3O_Y$ clean surface measured with different photon energy between 54 and 125 eV in comparison with MgKα XPS.

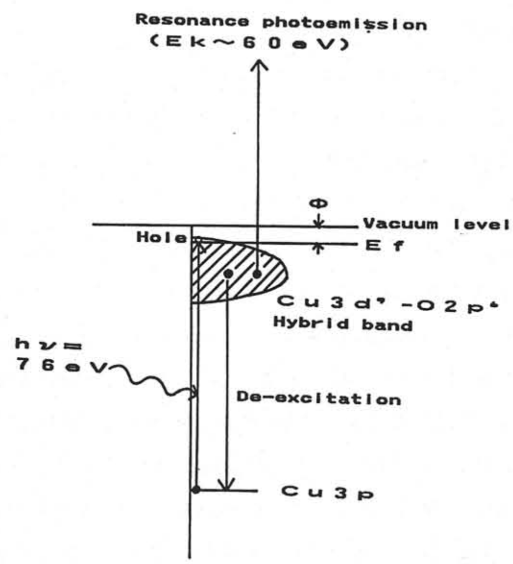

Fig.2 Mechanism of resonance photoemission from $YBa_2Cu_3O_Y$ superconductors at 76 eV of photon energy.

than the clean $YBa_2Cu_3O_Y$. Although no resonance peak was observed probably because of surface contamination, or oxygen deficient region at the as-received surface, the resonance peak was found to appear by O^+ ion sputtering for appropriate duration, that is for 10 min in this case. The O^+ ion incidence angle was set at 45 degree. Further O^+ ion sputtering was found to rather decrease the resonance peak. On the XPS Cu2p spectra, a shake-down type satellite peak which is characteristic of Cu^{2+} state also emerged. Although the resonance peak intensity was not recovered as high as that on the scraped surface, this dry technique is considered to be applicable to surface treatment prior to metal contact formation or other device fabrication processes.

Next, Au/$YBa_2Cu_3O_Y$ interfaces were analyzed using this resonance photoemission technique for contact formation. Au was chosen as a candidate for ohmic contact electrode, because it is a noble metal which would not react with $YBa_2Cu_3O_Y$. Even though up to nominally 30Å Au was deposited on the scraped $YBa_2Cu_3O_Y$ surface, the resonance peak was almost unchanged, which means that no interfacial reaction took place, as shown in Fig.4. Valence band features are changing from Cu3d-O2p hybrid band[6] to Au5d feature. Thus good contact properties are expected to be obtained by the combination of the O^+ ion sputtering and in situ Au deposition process. In fact, contact resistance of $5.5x10^{-7}\Omega cm^2$ at 77K[7] was obtained with the $YbBa_2Cu_3O_Y$ film. This is the lowest ever reported.

Finally, the junction formation process was investigated with amorphous Si deposition on a clean $YBa_2Cu_3O_Y$ surface. The resonance peak practically disappeared as shown in Fig. 5, when only about 1 monolayer of Si was deposited. At the first stage of Si deposition, a shoulder peak emerged at about 1.2eV higher B.E. than Cu3d-O2p hybrid band. This peak can be attributed to Si-O bonding. Judging from XPS Cu2p and Si2p peak intensities, photoionization cross sections and escape depths, the Si layer thickness at the fourth stage was calculated to be about 9 Å, by assuming a flat Si layer on $YBa_2Cu_3O_Y$ surface. The drastic change of resonance peak at the interface was also observed on Si-

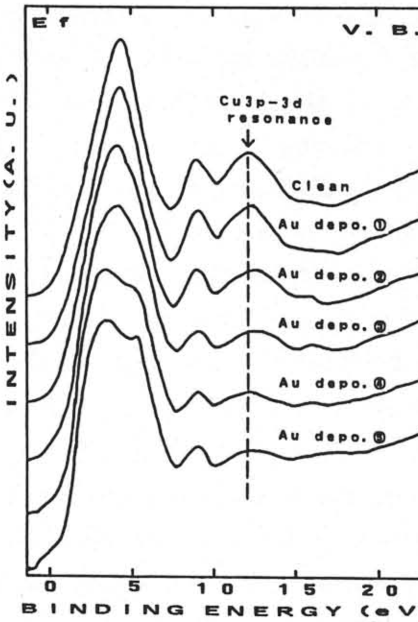

Fig.4 Valence band spectra from Au-covered $YBa_2Cu_3O_Y$ surfaces. $h\nu$=76 eV.

deposited surfaces of CuO which was formed by thermal annealing of a Cu sample in oxygen atmosphere. Thus, Si is found to easily be oxidized. Analogous phenomena were reported by D.M.Hill et al[1] in the case of Fe/LaSrCuO, where adatoms of Fe initially destroy the two dimensional character of the superconductor, withdrawing oxygen to form more energetically favorable Fe-O bonds.

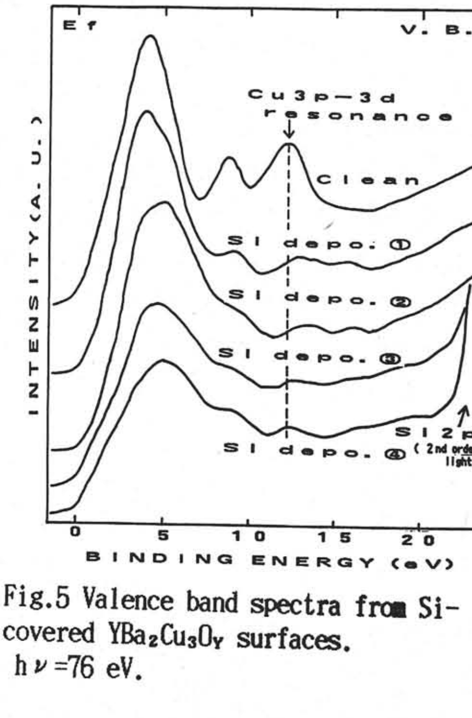

Fig.5 Valence band spectra from Si-covered $YBa_2Cu_3O_Y$ surfaces. $h\nu$=76 eV.

Figure 6 shows Si2p spectra from Si-deposited $YBa_2Cu_3O_Y$ surfaces. Si was in the SiO_x state at the interface. The XPS Cu2p peak also changed to the metallic state. Although it is difficult to determine whether the Cu2p is in the Cu state or Cu^{1+} state, we regard the reduced Cu as the Cu metallic state by comparing it with that in Cu metal. Thus strong interfacial reaction such as "Si + $Cu^{2+} \rightarrow Si^{2+}$(or Si^{x+}) + Cu" takes place even in the surface region detected by XPS. In other words, amorphous Si is not a good candidate for the junction material, because the XPS-detecting region which is deeper than the coherence length (5 - 20Å), is not in the superconducting state.

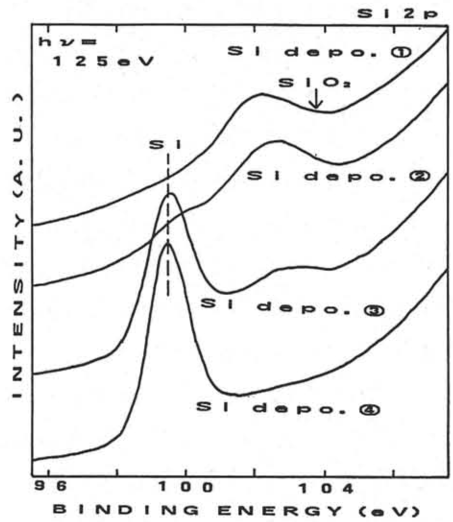

Fig.6 Si2p spectra from Si-covered $YBa_2Cu_3O_Y$ surfaces. $h\nu$=125 eV.

4. Conclusion

In conclusion, a practical dry cleaning procedure was developed to obtain very low contact resistance by using the Cu3p-3d resonance photoemission peak as a monitor of surface superconductivity. Furthermore in situ Au deposition on this cleaned surface is found to cause no interfacial reaction, and to provide a good ohmic contact. Amorphous Si deposition on $YBa_2Cu_3O_Y$ surface is not suitable for Josephson junction formation, judging from the SRPES results.

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