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Development of a cryostat X-ray fluorescence holography apparatus and its application

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1 Introduction

X-ray fluorescence holography (XFH)[1] is a relatively new local structural analysis method, which can determine atomic arrangements around a specific element without any prior knowledge of structures. It can directly reconstruct 3D atomic images using Fourier transformation. We consider that XFH and the related atomic resolution holographic techniques (photoelectron holography, neutron holography) are a third method of structural analysis at the atomic level after X-ray diffraction (XRD) and X-ray absorption fine structure (XAFS). As known by many researchers, XRD and XAFS are established methods that are widespread use in various fields. XRD and XAFS provide information on long-range translational periodicities and very local environments, respectively, whereas XFH gives 3D information on the local order and can visualize surrounding atoms with a large range of coordination shells. Such characteristics are major advantages of XFH.

XFH provides 3D atomic images within a diameter of several nm order. From the viewpoint of structural analysis, this novel feature of XFH has a strong possibility of opening new fields in solid-state physics. To date, using XFH we have obtained new information on the structures of some advanced materials. We call this feature of XFH “3D medium-range local structure observation”. Finding novel nanometer clusters is one of the major advantages of XFH. For example, in 2009, we studied the phase transition behavior of of $Ti_{50}Ni_{44}Fe_6$ single crystal, a shape-memory-alloy related material. This material exhibits parent (*P*), incommensurate (*IC*), and commensurate (*C*) phases when the temperature is decreased to below room temperature. While the atoms in $Ti_{50}Ni_{44}Fe_6$ are distributed homogeneously in the *P*-phase, they formed cluster-like structures with a radius of 8 Å in the *C*-phase, where the motion of the atoms inside the clusters is frozen. This is valuable information for understanding the phonon softening at the phase transition.[2]

To date, we have cooled samples by a liquid-nitrogen-spraying system. However, this system cannot cool samples below 100 K and cannot control sample temperatures with high accuracies. Therefore, we have not been able to study phase transition behaviours of superconductor materials, which often show interesting phenomena below 100 K, by measuring holograms. Thus, in this study, we developed a new XFH system with a cryostat, which can cool samples down to 5 K, and applied it to superconductor materials.

2 Experiment

Figures 1 (a) and (b) show the picture of the cryostat X-ray fluorescence holography apparatus and the schematic illustration, respectively. The apparatus consists of two axis goniometer and a cryostat. There is a piezoelectric rotation stage inside the cryostat as shown in Fig.1(b). We can know the angle of the piezoelectric stage by reading the resistance value of an conductor installed in the stage. XFH holograms are recorded by scanning the two axis goniometer and the piezoelectric stage and by measuring the angular distributions of the fluorescence intensities from samples.

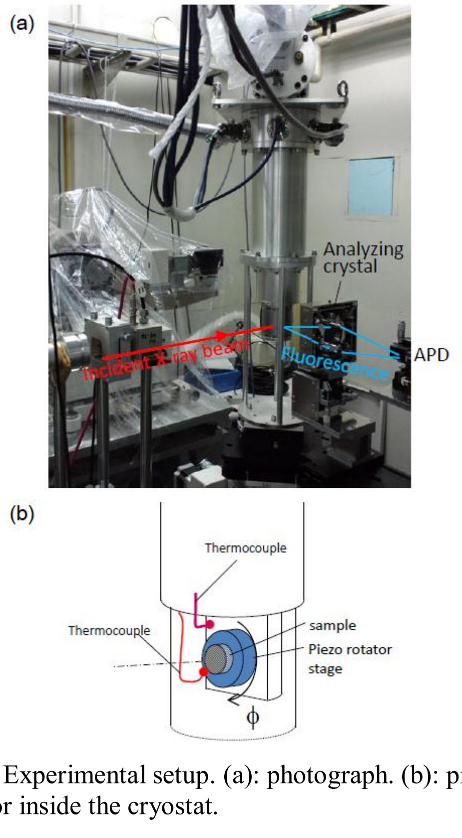

Fig.1 Experimental setup. (a): photograph. (b): piezo rotator inside the cryostat.

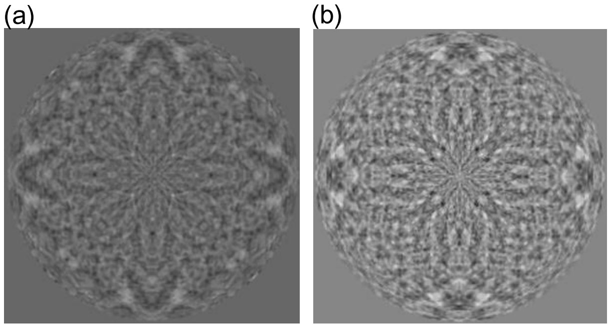

Fig.2 Cu Kα holograms of $La_{1.85}Sr_{0.15}CuO_4$ measured at (a) 50K and (b) 25K. The incident X-ray energy was 9.2 keV.

Here, we used $La_{1.85}Sr_{0.15}CuO_4$ as the measured sample. This material exhibits insulator-superconductor phase transition at 37.5 K. Therefore, we measured the Cu Kα holograms at the sample temperatures of 25 K and 50 K.

The X-ray beam from the bending magnet source was monochromatized by a Si(111) double-crystal monochromator and focused onto the sample with a Si bent total reflection mirror. The incident X-ray energies were 9.2 – 12.7 keV in 0.5 keV steps. Using the toroidally bent graphite crystal, Cu Kα fluorescent X-rays from the sample were analyzed and focused onto an avalanche photodiode. The fluorescence intensities were recorded by scanning the azimuthal angle of the sample ϕ in the range of $0° \leq \phi \leq 360°$ in 0.5° step and the incident angle θ in the range of $0° \leq \theta \leq 70°$ in 1° steps. The measurement time for one single-energy hologram was about 6 hours. The intensity of the each pixel was 200,000.

The lattice constants of $La_{1.85}Sr_{0.15}CuO_4$ are a = 5.3247 Å, b = 5.3486 Å, c = 13.1973 Å at 10 K, and a = 5.3266 Å, b = 5.3476 Å, c = 13.1992 Å at 70 K, and a = b = 5.3422 Å, c = 14.2317 Å at 295 K.

3 Results and Discussion

Figures 2 (a) and (b) show the measured holograms at 50 K and 25 K, respectively. The measured X-ray energy was 9.2 keV. The data were fourfold symmetrized using the crystal structure data of the sample. Both holograms exhibit some patters including X-ray standing wave lines. The difference between the two hologram patterns reflects the structural change by the superconductor phase transition at Tc = 37.5 K.

Figures 3 (a) and (b) show atomic images around Cu at 50 K and 25 K, respectively. Both images are distinctly different. The images at 50 K are weak and are little. The possible atomic image is marked by solid circle. This might correspond to 1st neighbor Cu image. But, other possible atomic images cannot be seen in Fig.3 (a). The images at 25 K are stronger and more than those at 50 K. The first neighbor Cu atomic image became stronger, and another possible Cu image marked by dashed circle appeared. This result indicates the occurrence of a long-range ordering below Tc = 37.5 K.

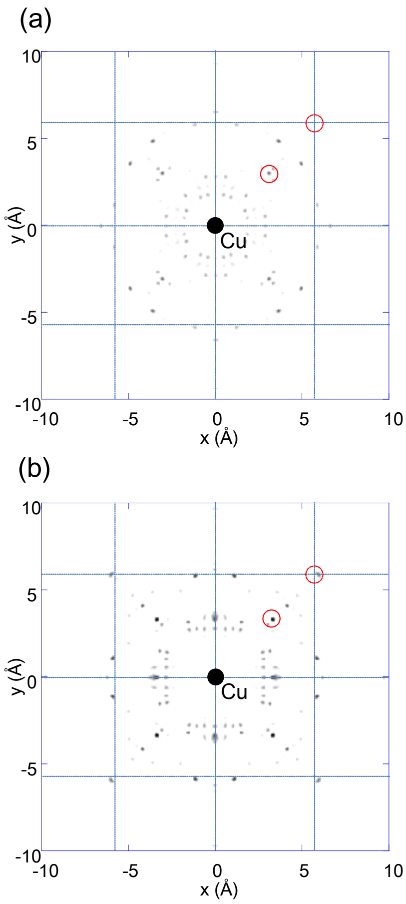

Fig.3 Atomic images of $La_{1.85}Sr_{0.15}CuO_4$ around Cu at z = 0.0 Å. (a): 50 K. (b):25K.

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