Electron distribution of hydrogen atom of squaric acid in high pressure phase

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1. Introduction

Squaric acid $C_4O_4H_2$ is one of the typical antiferroelectric material (P2/m) whose transition temperature to the high symmetry phase (I4/m) is 370K on raising temperature and 0.7GPa on raising pressure at room temperature.

Recently, high pressure Raman scattering experiments suggested that the nature of the hydrogen-bond changes from a double minimum potential to a single minimum potential at far above the transition pressure in the high symmetry phase associated with the construction of the bond length.

The purpose of the present work is that we would measure the electron distribution of hydrogen atoms in the hydrogen-bond at the high pressure phase directly by using X-ray diffraction. Within the short beam time, we will be able to test the capability of the structure analysis of a single crystal sample under high pressure at BL02B1. We also tested the program of the four-circle diffractometer and the imageprocessing program DENZO during the current beam-time.

2. Experimental

X-ray diffraction experiments were performed at the Crystal Structure Analysis beamline (BL02B1) in SPring-8. Double monochromators of Si 111 and two mirrors to eliminate the higher harmonics of the beam and to focus the beam were used. The energy of X-ray was 29.974keV.

3. Results and Discussion

We found that there were still program-bugs for the accumulation of the intensity data on the four-circle diffractometer mode. Because of this reason, we could not obtain the accurate angle origins of each axes. At the moment, we ignored the deviation of the absolute angles and determined the lattice parameters, as is tabulated in Table 1. The program-bug was fixed after this experiment. In the table, (a) is for the data of laboratory system with Mo Kα1, (b) for the vacuum camera at BL02B1, (c) for the data taken with the four-circle mode of BL02B1 at the ambient pressure and temperature. The data in (d) is taken from the sample in the diamond anvil cell with the four-circle mode of BL02B1. The pressure is 1.26GPa and it must be the high symmetry phase (a=c, tetragonal lattice). The length of axes seems good but there is deviation in lattice angles probably due to the uncertainty of the angle origin of the diffractometer.

We performed structure analysis. For (a), R-factor is 3.9% by using 1081 data points. Concerning the data for (b), the number of the picked-up data by DENZO is 15049, and the preliminary analysis by using $I>10\sigma(I)$ data, whose number is about 4000-point, gives the R-factor 5.6%. We measured 175 data points for (d) at $2\theta<30$. By looking the profile, we picked up 40 data points. The structure analysis is not finished yet because there are contamination from diamond and gasket or beryllium. Typical intensity for (d) is that the Bragg reflection (2 -1 -1) from the sample is 120kcps at the peak, 12 Mcps from the diamond and 10kcps from the gasket or beryllium.

	(a)	(b)	(c)	(d)
a	6.132(1)	6.130(6)	6.126(3)	6.078(8)
b	5.273(1)	5.273(4)	5.278(3)	5.135(4)
c	6.145(1)	6.142(7)	6.144(3)	6.079(2)
α	89.99(2)	90.00(9)	90.15(5)	90.16(6)
β	89.95(1)	90.05(9)	89.66(5)	89.99(8)
γ	89.98(2)	90.00(8)	89.84(5)	89.89(9)
V	198.7(1)	198.5(3)	198.4(2)	189.7(3)

Table1 Obtained lattice parameters. (a) laboratory system, (b) vacuum camera, (c) four-circle mode at ambient pressure and temperature, (d) four-circle mode at 1.26GPa.