Crystal Structure Analysis of the Fullerene Compounds by the Maximum Entropy Method

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It is well known that many alkali-dope fullerenes shows superconductivity at relatively high temperature. In order to understand such an important physical property, it would be extremely interesting to know the electronic level structure of these compounds. The Maximum Entropy Method (MEM) is most appropriate method for the present purpose because it can reveal detailed electron density distributions of the crystalline materials directly from the experimental data without using any structural model.

The experiment were carried out at BL-02B1 by using IP as a detector. The Rb_2CsC_{60} specimen are sealed into 0.1mm silica glass capillary. The wavelength of incident X-ray was 0.497 A with 0.5mm beam size. The exposure time was 2.5 hrs. Since all of the integrated Bragg intensities are measured, any instability of incident X-rays shall not give serious effects onto the measured Bragg intensities in this experimental arrangement. This is a big advantage of the present method over the conventional point by point step scan. The maximum 2θ was about 25^0 which is not big. But because of very short wavelength of incident X-rays, the resolution in real space expressed by the minimum d-spacing is still 1.1 A which is not too bad for the crystal analysis of fullerene compounds.

As pointed out in the previous report (1997B0010-ND-np), the drawback of using very short wavelength as incident X-ray, i.e. much smaller peak separations in diffraction were again overcome by the extreme parallel beam of SPring-8 SR light.

The observed intensities were analyzed by the Rietveld analysis first. This is now rather common approach of the newly developed analytical method, that is, the combination of Rietveld and MEM. In the present report, only the results of Rietveld method shall be included.

In the Rietveld method, it is necessary to construct crystal structure model. First, the crystal model which assume C_{60} cage is spherical shell, which is very appropriate for pure C_{60}, because C_{60} molecules in pure C_{60} crystal shows almost free rotation at ambient temperature. However this model was not successful for Rb_2CsC_{60}. Then the merohedral disorder model was tried. It turned out that this crystal model gives fairly low R-factor, i.e. R_I=6.4%. Some other R-factors are R_P=2.6%, R_{WP}=3.3% and R_F=6.1%. The results of the Rietveld analysis are shown in Fig. 1. It is concluded that the basic structure of Rb_2CsC_{60} is merohedral disorder which means the rotation of C_{60} molecules is prohibited even at ambient.

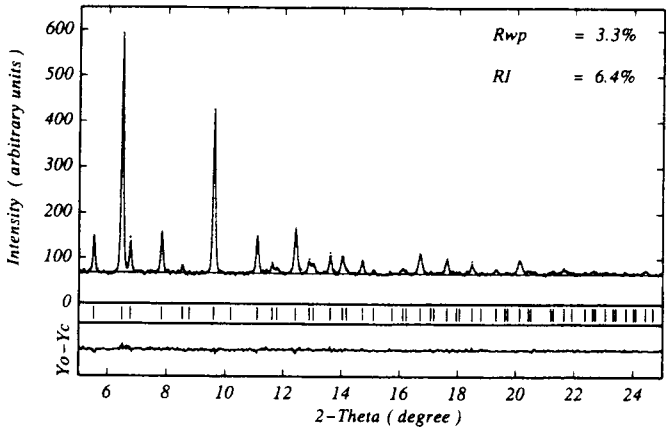

Fig.1. The Rietveld analysis for Rb_2CsC_{60}.