

these results, we can conclude that the density decrease of fluid Hg is essentially caused by the reduction of the coordination number through the entire density region as seen in the behavior of  $N_B$ . The variation in  $N_A$  gives more detailed information about the structural change accompanied by the M-NM transition.  $N_A$  represents the coordination number at the shortest distance in the first coordination shell, so the density variation of  $N_A$  suggests that the change in the nearest part of the first coordination shell is strongly related to the M-NM transition. As the most important observation, the gross feature of the density variation in  $N_A$  and  $r_1$  in [Figure 3](#) suggests that there exist three different regions in the density: the metallic region from 13.6 to about 10 g/cm<sup>3</sup>, the M-NM transition region from 10 to the critical density of about 6 g/cm<sup>3</sup> and the dense vapor region.

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## CRYSTAL STRUCTURE ANALYSIS OF THE FULLERENE COMPOUNDS BY THE MAXIMUM ENTROPY METHOD

Alkali metal doped fullerenes in the form of  $A_2BC_{60}$  are fascinating substances. Many of them show superconductivity while a few others show no superconductivity. As the fundamental crystal structure, it is well known that  $C_{60}$  molecules form an fcc lattice and alkali metal atoms locate at both the tetrahedral and octahedral sites [1]. There is a close relation between the lattice constant of the compounds and their superconducting transition temperature,  $T_c$  [2,3]. For example,  $Rb_2CsC_{60}$  has a relatively high  $T_c$ . On the other hand,  $Li_2CsC_{60}$  shows no superconductivity.

It has been suggested that different bonding natures exist depending on their lattice constants and that they are closely related to the superconducting property [4]. Bonding nature may be divided into two regions, *i.e.*, the interatomic region between the doped metal atoms and the carbon-carbon region on  $C_{60}$  molecules. So far, there has been no definite experimental evidence on bonding nature. In this study, at the beamline **BL02B1**, the fine structure of  $Rb_2CsC_{60}$  and  $Li_2CsC_{60}$  are revealed [5], including the bonding nature for alkali metal doped fullerenes by the Maximum Entropy Method (MEM), which is an advanced imaging technique using diffraction data [6].

The MEM charge densities of  $Rb_2CsC_{60}$  and  $Li_2CsC_{60}$  are shown for a (110) plane in [Figures 1\(a\)](#) and [\(b\)](#), respectively. At a glance, it can be easily seen that there are distinct structural differences between  $Rb_2CsC_{60}$  and  $Li_2CsC_{60}$ . A non-superconducting alkali metal doped fullerene,  $Li_2CsC_{60}$ , has uniform charge densities of the  $C_{60}$  molecule due to nearly free rotation of  $C_{60}$ . In contrast, a superconducting alkali metal doped fullerene,  $Rb_2CsC_{60}$ , has some kinds of disorder. To visualize three-dimensional distributions of the charge on the carbon cage in  $Rb_2CsC_{60}$ , the MEM electron density distribution of  $C_{60}$  and Rb atoms are shown by an equi-contour surface at  $2.0 \text{ e } \text{Å}^{-3}$  in [Figures 2\(a\)](#). In this figure, the characteristic features of the merohedral disorder, the hexagons facing toward Rb atoms and cloverleaf features,

be clearly seen. For reference, a schematic model of merohedral disorder is shown in Figure 2 (b). In order to show a strong structural contrast between  $Rb_2CsC_{60}$  and  $Li_2CsC_{60}$ , their three-dimensional equi-contour surfaces are shown in a Fig. 2 (a) and Figure 3, respectively.

In Figure 3, there are no localized densities in the interatomic region for these materials. Therefore, the chemical bond nature of doped alkali metals is ionic, which is independent of the existence of superconductivity. The electronic charges of doped alkali metals were estimated from the amount of electrons around a certain atomic site.

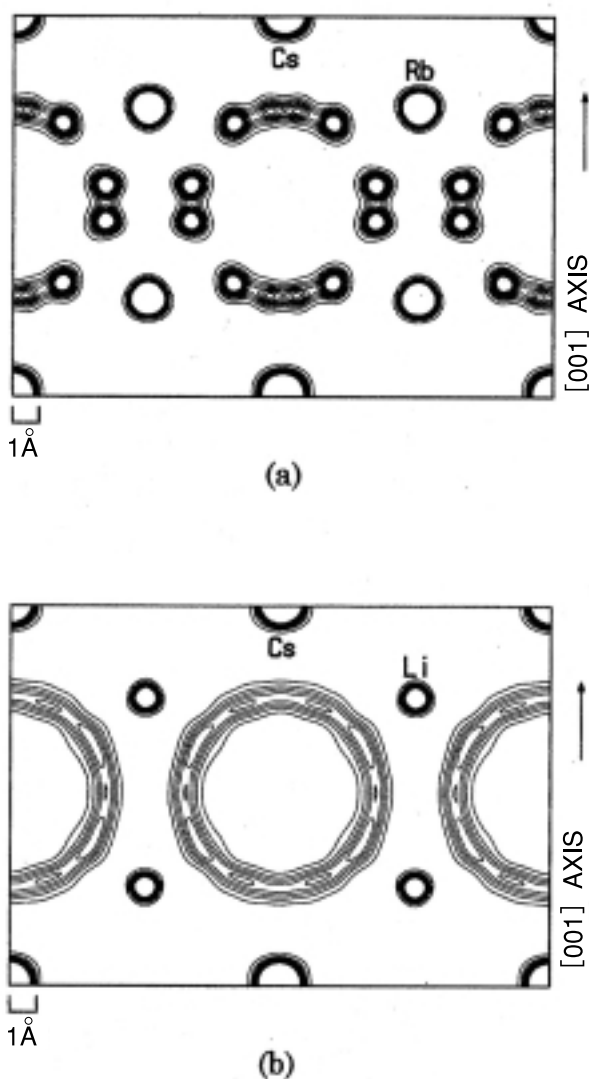


Fig. 1: (110) plane MEM charge densities of (a)  $Rb_2CsC_{60}$  and (b)  $Li_2CsC_{60}$ . Contour lines are drawn from 0.0 to 5.0  $e \text{ \AA}^{-3}$  with 0.5  $e \text{ \AA}^{-3}$  intervals.

The results are shown in Table I. Since it is well known that there is significant site exchange between the octahedral and tetrahedral sites for the present materials, a deficiency of charges at each site must be affected by the site exchange for each material. By taking possible site exchanges into account, it should be noted that these values cannot be considered the absolute valence of the atoms. However, it is evident that the total value, which must be a much more reliable value than that of each site, decreases as  $T_c$  decreases. This implies the charge transfer from the doped metal atoms to the  $C_{60}$  molecule could be a crucial factor for creation of superconducting fullerides. From the crystal structural point of view, the difference in the strength of interaction between the metal atoms and the  $C_{60}$  molecule seems to cause the difference for the disordered state of the  $C_{60}$  cage in a crystal, i.e., the much stronger interaction between alkali metal atoms and  $C_{60}$  prevents free rotation of the  $C_{60}$  cage in  $Rb_2CsC_{60}$ . In the present electron level structural study, it was found that there are distinct structural differences relating to the superconducting

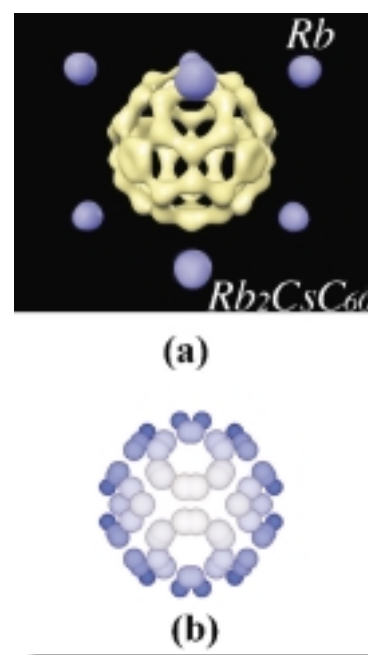
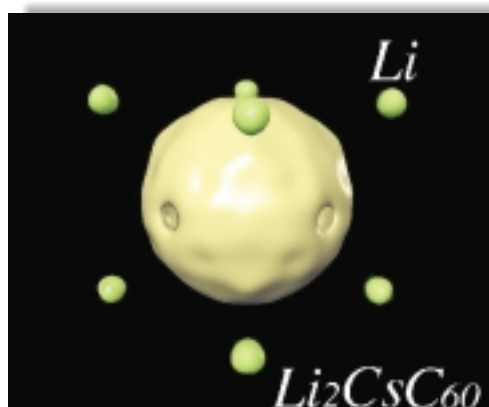


Fig. 2: (a) Equi-contour ( $2.0 e \text{ \AA}^{-3}$ ) density map for the MEM charge densities for  $Rb_2CsC_{60}$ . (b) Merohedral disorder model of  $C_{60}$ .

**Table 1** . Deficiency of charge at tetrahedral, octahedral sites and the total.

	Li <sub>2</sub> CsC <sub>60</sub>	Rb <sub>2</sub> CsC <sub>60</sub>
Tetrahedral site	0.3(1) e	2.1(2) e
Octahedral site	0.2(1) e	2.0(2) e
<b>Total value</b>	<b>0.5(2) e</b>	<b>4.1(4) e</b>



**Fig. 3:** Three-dimensional equi-contour (2.0 e Å<sup>-3</sup>) density map of Li<sub>2</sub>CsC<sub>60</sub> .

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## NUCLEAR RESONANT INELASTIC AND QUASI-ELASTIC SCATTERING EXPERIMENTS

Nuclear resonant inelastic scattering has many interesting features, and one of the most important is that it is possible to excite a specific nuclide [1]. For the compounds composed of more than one element, not all of the atomic motions are the same, and they depend on their mass differences, binding states and atomic positions. Furthermore, in a solution that contains different kinds of ions, the motion of each ion is different. For atomic motions limited to one- or two-dimensional regions, it is desirable to observe only their intrinsic dynamics without the influence of the surroundings. The nuclear resonant scattering method is effective for these areas of research and is expected to be a new tool for the study of dynamics of atoms. We have measured nuclear resonant inelastic and quasi-elastic scattering for several samples at the beamline **BL09XU**. Here, we introduce two examples of these.

Graphite is a typical layered material, which intercalates various atoms. Intercalated graphites show various stage structures depending on the sample preparations. We synthesized stage-1 graphite-ferric-chloride-intercalation compounds and measured the phonon energy spectra [2]. Our purpose is to study the dynamics of ferric chloride confined in the layers by observing the phonon densities of states projected to the layer direction and its vertical direction [3]. Energy spectra of nuclear resonant inelastic scattering were measured for two angles ( 0° and 90° ) between the direction of incident X-ray and the normal to the graphite layers.

The experimental setup is shown in Figure 1, and the observed spectra are shown in Figure 2. When the incident X-ray direction is parallel to the graphite layers, phonon modes with parallel components to the layers are excited. In case the incident X-ray direction is perpendicular to the graphite layers, phonon modes with perpendicular components to the layers are excited. The observed phonon modes parallel and perpendicular to the layers increase below 5 meV and around 10 meV, respectively. That is, the phonon modes