 MLF Experimental Report	提出日 Date of Report July 4, 2013
課題番号 Project No. 2012B0243 実験課題名 Title of experiment Studies on phonon modes reflecting ferroelectricity in a charge transfer complex β' -(BEDT-TTF) ₂ ICl ₂ s 実験責任者名 Name of principal investigator Seiko Kawamura 所属 Affiliation MLF, J-PARC	装置責任者 Name of responsible person Kenji Nakajima 装置名 Name of Instrument/(BL No.) AMATERAS (BL14) 実施日 Date of Experiment February 25 - March 2, 2013

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。(適宜、図表添付のこと)
 Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.
Name of sample: β' -(BEDT-TTF) ₂ ICl ₂ Formula: (C ₁₀ S ₈ H ₈) ₂ ICl ₂ State and weight: tiny single crystals, 1.5 g

2. 実験方法及び結果 (実験がうまくいかなかった場合、その理由を記述してください。) Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.
<p>Experimental method</p> <p>β'-(BEDT-TTF)₂ICl₂ is an organic dimer-Mott insulator having a quasi-two dimensional (2D) crystal structure. About 230 pieces of tiny single crystals are aligned on aluminum plates so that the a^*-axis is perpendicular to the aluminum plate (namely the 2D plane is parallel to the plate) and that the c-axis is co-aligned with each other (Fig. 1). The aluminum plates on which the samples were mounted were set to an aluminum frame to set into a standard aluminum sample cell together with helium gas (Fig. 1). First we set the sample so that both the a^* and b^* axes are within the horizontal plane to collect the data for $Q // a^*$ and $Q // b^*$, and next we changed the configuration so that the a^* and c^* axes are within the horizontal plane to collect the data for $Q // c^*$. A closed-cycle ⁴He refrigerator was used to cool the sample. The incident neutron energies $E_i = 42.1, 15.2$ and 7.7 meV were chosen.</p> <div data-bbox="981 1702 1428 2027" data-label="Image"> </div> <p style="text-align: right;">Fig. 1</p>

2. 実験方法及び結果(つづき) Experimental method and results (continued)

Results

We have studied phonon modes reflecting spin and charge degrees of freedom in this system (Project use at BL14, No. 2012P0202), and then we found that the intensity of the optical mode at $E = 4.2$ meV is enhanced at a zone boundary at a glassy ferroelectric transition ($T_{FE} = 62$ K) and further at a long-range antiferromagnetic transition ($T_N = 22$ K). However, we could not reveal clearly that this mode is coupled with the charge and spin because of poor data points (only three points: 7.5, 35 and 70 K). In the present study, we added some temperature points. Figure 1(a) shows the obtained E - Q slice along the k direction at 80 K. Here a limited region around $h \sim 0$ and $l \sim 0$ to 1 was integrated to extract information at a zone boundary (0, 5.5, 0). The temperature dependence of the intensity for the 4.2 meV-mode at a zone boundary, which is indicated by a square in Fig. 1(a), is shown in Fig. 1(b). A Bose factor is already corrected. Open circles are the previous data, and closed circles are the present data normalized with the intensity at 7.5 K. The enhancements of this intensity at T_{FE} and further at T_N were clearly observed.

Furthermore, we investigated anisotropy of the phonon intensity by collecting data for $Q // a^*$, $Q // b^*$ and $Q // c^*$. Figures 3(a), 3(b) and 3(c) show their E - Q slices, where the whole measured regions of the two other axes are integrated. A Bose factor is already corrected. The energy-cuts at $h = 5.5 \pm 0.05$, $k = 4.5 \pm 0.05$ and $l = 2.5 \pm 0.05$ are shown in Figs. 3(d), 3(e) and 3(f), respectively. The a^* -axis is perpendicular to the 2D plane, and the b^* -axis roughly corresponds to the direction in which charge disproportionation is expected to occur below T_{FE} . Several optical modes were observed in each Q direction. Their intensity shows similar temperature dependence, while it seems to depend on the direction. Although mode assignment for this system is quite difficult, the observed axis dependence may give some information on the vibration modes in this system. More analysis will be continued.

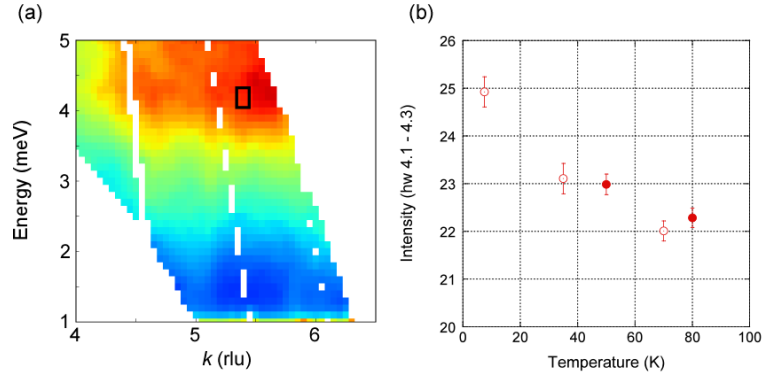


Fig. 2: Results of the measurements for $Q // b^*$. (a) E - Q slice for the thin h and l region at 80 K. (b) Temperature dependence of the intensity for $k = 5.5 \pm 0.05$ and $E = 4.2 \pm 0.1$ meV, which is indicated by a square in (a). Data obtained in the previous (June 2012) and present studies are plotted with open and closed circles, respectively.

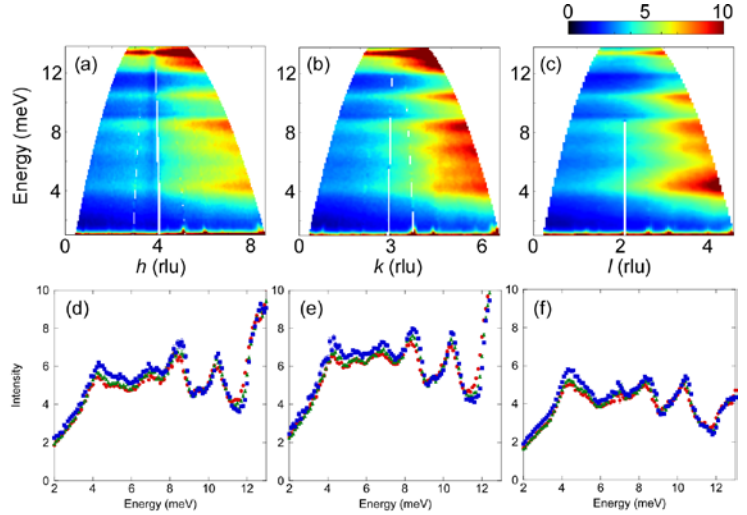


Fig. 3: E - Q slices for (a) $Q // a^*$, (b) $Q // b^*$ and (c) $Q // c^*$ at 7.5 K. Energy cuts at (d) $h = 5.5 \pm 0.05$ obtained from (a), (e) $k = 4.5 \pm 0.05$ from (b), and (f) $l = 2.5 \pm 0.05$ from (c), observed at 80 K (red), 50 K (green) and 7.5 K (blue).