 <b>MLF Experimental Report</b>	提出日 Date of Report 2009/07/06
課題番号 Project No. 2008A0021 実験課題名 Title of experiment $\mu$ SR study of organic antiferromagnet $\beta'$ -(BEDT-TTF) <sub>2</sub> IBrCl 実験責任者名 Name of principal investigator Kazuhiko Satoh 所属 Affiliation Graduate School of Science and Engineering, Saitama University	装置責任者 Name of responsible person Yasuhiro Miyake 装置名 Name of Instrument/(BL No.) D1 実施日 Date of Experiment 2009/02/11 ~ 2009/02/14

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.
$\beta'$ -(BEDT-TTF) <sub>2</sub> IBrCl

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>Among the organic systems, superconductivity with the highest transition temperature is realized in <math>\beta'</math>-(BEDT-TTF)<sub>2</sub>ICl<sub>2</sub> [1]. At ambient pressure, <math>\beta'</math>-(BEDT-TTF)<sub>2</sub>ICl<sub>2</sub> shows semiconducting behavior and antiferromagnetic transition at <math>T_N = 22</math> K [2, 3]. As one-dimensional character of the Fermi surface is suggested from the band structure calculation [4], it is considered that the insulating behavior in <math>\beta'</math>-(BEDT-TTF)<sub>2</sub>ICl<sub>2</sub> is due to the strong electron-correlation effect. We have carried out zero-field <math>\mu</math>SR measurements for <math>\beta'</math>-(BEDT-TTF)<sub>2</sub>ICl<sub>2</sub> and detected spontaneous precession signal in the antiferromagnetic state [5]. We also found that the <math>\beta'</math>-(BEDT-TTF)<sub>2</sub>ICl<sub>2</sub> can be metallized by the application of 8-GPa pressure and shows the superconducting transition, <math>T_c</math>, at 14.2 K [1]. This is the highest superconducting transition temperature among organic superconductors to date. Origin of superconductivity is not clear at present, but antiferromagnetic correlation is considered to play an important role from the similarity of the temperature-pressure phase diagram to those of the well-known <math>\kappa</math>-phase superconductors.</p>

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

In order to understand the nature of superconductivity of  $\beta'$ -(BEDT-TTF)<sub>2</sub>ICl<sub>2</sub>, it is interesting to study related materials. We carried out muon spin relaxation measurements for isostructural compound  $\beta'$ -(BEDT-TTF)<sub>2</sub>IBrCl. This salt is also semiconducting antiferromagnet with  $T_N = 19.5$  K and shows a sign of superconducting transition at 8 GPa with  $T_c = 7.2$  K [6]. Single crystals of  $\beta'$ -(BEDT-TTF)<sub>2</sub>IBrCl were grown by electrochemical oxidation of BEDT-TTF, using (n-Bu)<sub>4</sub>NIBrCl in tetrahydrofuran. Zero- and longitudinal-field  $\mu$ SR experiments down to 3.2 K were carried out at D1 port of MLF, J-PARC.

Figure 1 shows zero-field  $\mu$ SR spectra at paramagnetic and antiferromagnetic states. Spectra in the paramagnetic state are well fitted by the sum of the temperature-independent static Kubo-Toyabe function and time- and temperature-independent Ag background  $A_{Ag}$ . Spontaneous muon spin precession signal is observed below the  $T_N$  suggesting the bulk nature of the antiferromagnetic state. Spectra in the antiferromagnetic state were fitted by the following formula;

$$A(t) = A_1 \exp(-\lambda t) \cos(2\pi f t + \alpha) + A_{Ag}.$$

As shown in Fig. 1, about 3 % of initial asymmetry disappears below the Néel temperature. Temperature dependence of the precession frequency  $f$  is shown in Fig. 2. The precession frequency gradually decreases toward  $T_N$ . It is found that  $\mu$ SR spectra and temperature dependence of precession frequency for  $\beta'$ -(BEDT-TTF)<sub>2</sub>IBrCl are similar to those for  $\beta'$ -(BEDT-TTF)<sub>2</sub>ICl<sub>2</sub>. In order to clarify the relation between magnetism and superconductivity in these salts, further  $\mu$ SR experiments under high pressure are desired.

references

- [1] H. Taniguchi *et al.*, J. Phys. Soc. Jpn., 72 (2003) 468.
- [2] M. Tokumoto *et al.*, Synth. Met. 19 (1987) 215.
- [3] N. Yoneyama *et al.*, Synth. Met. 86 (1997) 2029.
- [4] H. Kobayashi *et al.*, Chem. Lett. 15 (1986) 89.
- [5] K. Satoh *et al.*, Physica B374-375 (2006) 99.
- [6] K. Uchiyama *et al.*, J. Phys. IV France 114 (2004) 387.

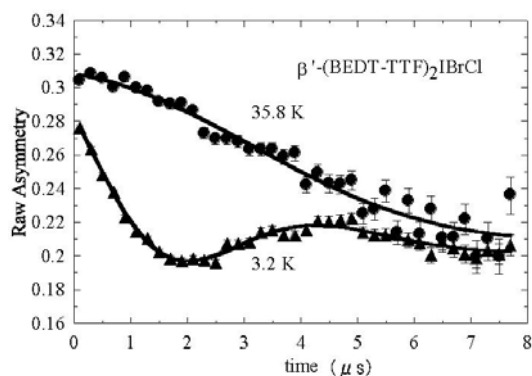


Fig. 1 Zero-field  $\mu$ SR spectra for  $\beta'$ -(BEDT-TTF)<sub>2</sub>IBrCl.

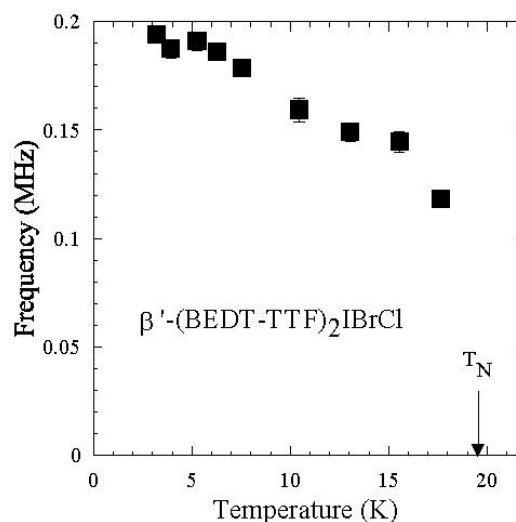


Fig. 2 Temperature dependence muon precession frequency for  $\beta'$ -(BEDT-TTF)<sub>2</sub>IBrCl.