Anomalous antiferromagnetic resonance of $\lambda$-(BETS)$_2$FeCl$_4$ in antiferromagnetic insulating phase

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The $\pi$-$d$ molecular conductor $\lambda$-(BETS)$_2$FeCl$_4$ consists of the conducting $\pi$-electrons in BETS molecules and the magnetic $d$-electrons in FeCl$_4^-$, Thanks to the strong interaction between $\pi$- and $d$-electrons, $\lambda$-(BETS)$_2$FeCl$_4$ has a fascinating temperature($T$)–magnetic field($B$) phase diagram including a paramagnetic metal phase, an antiferromagnetic insulating (AFI) phase and a field-induced superconducting phase [1-2]. However, the ground state of the AFI phase is still under debate [2]. It was previously believed that the AFI phase is due to the antiferromagnetic long-range order of the $d$-electrons. However, recent heat capacity measurement observed a Schottky anomaly below $T_N$, and its origin was explained from the contribution of paramagnetic $d$-electrons. This suggests that the $d$-electron remains paramagnetic below $T_N$, and only the $\pi$-electron becomes antiferromagnetic and localized [3]. Nevertheless, this explanation is not consistent with the existence of strong $\pi$-$d$ interaction, and contradicts with previous electron spin resonance (ESR) studies [4-6]. In addition, anomalous dielectric behavior due to the metastable state of the $\pi$-electrons was reported for the AFI phase [4]. Hence, to have a more microscopic information of the AFI phase, we are studying $\lambda$-(BETS)$_2$FeCl$_4$ using ESR spectroscopy.

We previously presented [7] that the easy-axis of $\lambda$-(BETS)$_2$FeCl$_4$ is changing with temperature. In order to study the relation between the change of the easy-axis and the external field, we have investigated the field-dependence of antiferromagnetic resonance (AFMR) using high-field ESR spectroscopy. When the magnetic field is applied parallel to the easy-axis, the easy-axis mode of AFMR is observed below 5 T, however, the easy-axis mode of AFMR changes to the hard-axis mode of AFMR above 5 T as shown Fig. 1 (conventional AFMR mode is shown in the inset of Fig.1). This suggests the change of the easy-axis occurs above 5 T. In addition, the change of permittivity was reported around 4–6 T [4], which suggests that the magnetic properties of $\lambda$-(BETS)$_2$FeCl$_4$ should be strongly connected with the electronic state of the $\pi$-electrons. Therefore, the change of the easy-axis is supposed to be due to the metastable state of the $\pi$-electrons. Moreover, splitting of AFMR signals is observed above 5,9 T (as shown Fig.1), this behavior suggests two different environments of Fe spins exist.

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Fig. 1: Frequency-field dependence of AFMR ($B$//easy-axis, $T = 2$ K) The inset shows frequency-field dependence of conventional AFMR