Antiferromagnetic Insulating Phase of λ-$(BETS)_2FeCl_4$ Studied by Electron Spin Resonance

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Molecular conductors with finite π-*d* interaction have attracted considerable interests for the past few decades since interaction between the itinerant π-electron and localized *d*-electron yields some intriguing physical phenomena. λ-$(BETS)_2FeCl_4$ is one of the such materials which shows interesting physical properties. Although it is well-known that this salt shows superconducting state above 17 T when the magnetic field is applied along the conducting plane [1], the ground state at low temperature is also interesting. The high-temperature phase of this system is paramagnetic and metallic (hereafter, the PM phase). Then, this salt shows a metal-insulator (MI) transition, and becomes simultaneously antiferromagnetic at T_{MI} = 8.3 K known as the antiferromagnetic insulating (AFI) phase. The origin of the antiferromagnetic state, whether the *d*-electrons or the π-electrons trigger the antiferromagnetic state, is discussed for a long time. However, recent specific heat measurements revealed that the broad excess specific heat observed below T_{MI} can be fit with the Schottky peak in which the six energy levels of Fe^{3+} (S=5/2) is split by an internal field of 4 T. Therefore, it is now considered that only π-electrons become antiferromagnetic (inducing an internal field of 4 T), and the *d*-electrons of Fe^{3+} remain paramagnetic [2]. However, previous ESR studies did not observe electron paramagnetic resonance (EPR) in the AFI phase, and only antiferromagnetic resonance (AFMR) was reported. [3-7] Meanwhile, many dielectric anomalies are reported nearby the phase boundary of the PM and AFI phases [3,6]. We think that the observation of dielectric anomalies just below T_{MI} are due to the non-localized π-electrons, which affect the magnetism of the system, and are the origin of the excess specific heat. Hence, to resolve these issues, we are performing ESR measurements on λ-$(BETS)_2FeCl_4$, and studying its angular and temperature dependences in detail.

Last fiscal year, we have studied the EPR of λ-$(BETS)_2FeCl_4$ in the PM phase, and reported that the angular dependence has some characteristic features owing to the strong π-*d* interaction [7]. Moreover, we have proposed a method to find the antiferromagnetic easy-axis from the angular dependence of EPR. This fiscal year, we have applied the magnetic field along the easy-axis by using this method, and studied the AFMR in detail.

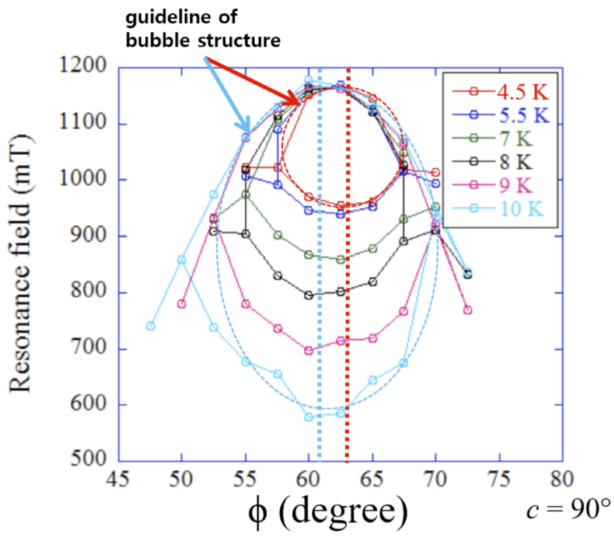

Figure 1 Angular dependence of AFMR near the easy-axis.

Figure 1 is the angular dependence of AFMR using the X-band ESR system. In general, a bubble structure, which consists of spin-flop resonance and the easy-axis mode of AFMR, is observed for a low-frequency region such as the X-band when the magnetic field is applied nearby the antiferromagnetic easy-axis. The center of the bubble corresponds to the easy-axis, and the top and the bottom part of the bubble corresponds to the spin-flop resonance and the easy-axis mode of AFMR, respectively.

In consistency with the previous report, the spin-flop resonance is observed around 1150 mT (top part of the bubble) [8]. Moreover, the easy-axis mode of AFMR shifts to lower field as the temperature increases, which is also consistent with the previous report [4-6]. However, we have observed for the first time that the easy-axis (i.e. center of the bubble) tilts as a function of temperature.

Anomalous AFMR signals are also observed in the high magnetic field region. Although the frequency dependence of AFMR shows a conventional easy-axis mode of AFMR, the AFMR signal splits above 6 T (see open and solid triangles in Fig. 2). The split of AFMR suggests a transition to a different ground state, and it seems to be an intrinsic behavior since the signal split is also observed for *B*//*a*- and *B*//*c*-axes as shown in Fig. 3. Moreover, the easy-axis mode seems to change to the hard-axis mode for the *B*//*c*-axis, and vice versa for the *B*//*a*-axis above 4 T as shown in Fig. 3.

In summary, the AFMR below 5 K shows the typical behavior of the antiferromagnet with uniaxial anisotropy. However, interesting behavior, including the tilt of the easy-axis and the AFMR signal spilt, has been observed near the boundary between the AFI and PM phases. It is supposed that the dielectric anomalies observed at the phase boundary are the origin of the observed anomalous antiferromagnetic state.

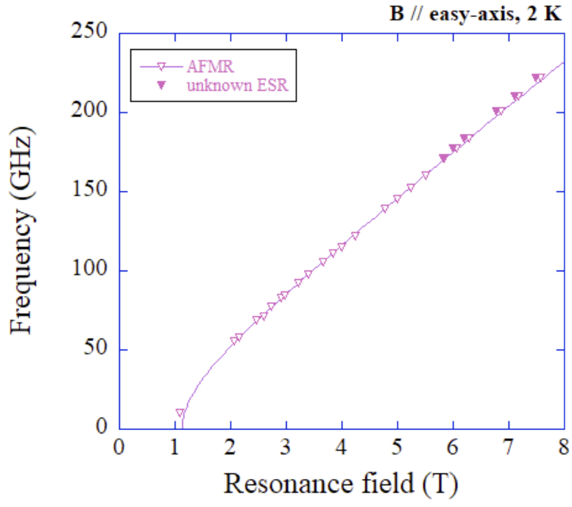

Figure 2 Frequency-resonance field plots of AFMR for *B* // easy-axis at 2 K

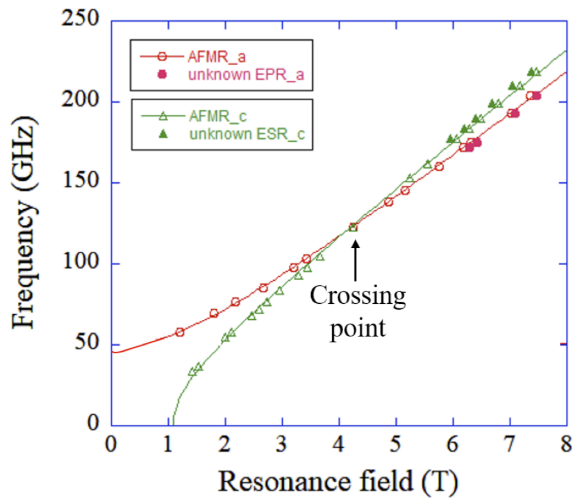

Figure 3 The frequency dependence of the resonance field for *B*//*c*- and *a*-axes.

References
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