High field ESR measurements of Me₄As[Pd(dmit)₂]₂

H. Ohta a, b, *, T. Sakurai a, S. Okubo a, b, R. Kato c, T. Nakamura d

a Department of Physics, Kobe University, 1-1 Rokkodai, Nada, Kobe 657-8501, Japan
b Venture Business Laboratory, Kobe University, 1-1 Rokkodai, Nada, Kobe 657-8501, Japan
c The Institute of Physical and Chemical Research, Wako, Saitama 350-0198, Japan
d Institute of Molecular Science, Myodaiji, Okazaki, 444-8585, Japan

Abstract

High field ESR measurements of Mott-Hubbard insulator Me₄As[Pd(dmit)₂]₂ have been performed in the frequency region from 50 to 315 GHz using the pulsed magnetic field up to 15 T. Broad and sharp resonances were observed in the temperature region from 1.8 to 77 K. The origins of these resonances were discussed from the temperature and frequency dependences in connection with the antiferromagnetic order suggested by H-NMR at 35 K.

Keywords: Electron spin resonance, Metal-insulator phase transition, Magnetic phase transition, Organic conductors based on radical cation and/or anion salts

Pd(dmit)₂ compounds are very interesting system with the one dimensional LUMO (bonding) band located below the two dimensional HOMO (anti-bonding) band. They are Mott-Hubbard insulators under ambient pressure but the application of pressure induces an overlap of these two bands and changes the physical properties [1]. For instance, β-Me₄N[Pd(dmit)₂]₄ is a high pressure superconductor (Tc=6.2 K at 6.5 kbar [1]) while this salt shows no clear metallic behavior in the low pressure region. β'-Me₄As[Pd(dmit)₂]₂ is one of these series of compounds and it undergoes an antiferromagnetic order below TN=35 K with a small local magnetization from H-NMR study [3]. X-band ESR was difficult in the high temperature region due to the broad linewidth and no ESR was observed below TN in the X-band ESR measurement [4]. The aim of this paper is to study the antiferromagnetic state of β'-Me₄As[Pd(dmit)₂]₂ by extending the frequency and magnetic field regions with our high field ESR.

High field ESR measurements have been performed in the frequency region from 50 to 315 GHz using Gunn oscillators. The temperature region is from 1.8 to 80 K and the pulsed magnetic field up to 15 T is used. The details of our experimental setup can be found in refs. [5, 6]. About 100 needle like single crystals of Me₄As[Pd(dmit)₂]₂ were aligned on the polyethylene sheet using GE varnish in order to gain intensity of ESR.

Figure 1 shows the typical temperature dependence of Me₄As[Pd(dmit)₂]₂ observed at 160 GHz for H//a*. Two broad and sharp absorption lines around 6 T are observed in the whole temperature region between 4.2 K and 77.7 K. The temperature dependence of the linewidth of the sharp absorption line observed in Fig. 1 is shown in Fig. 2. It is clear that the broadening of the linewidth occurs at TN. Together with the fact that the g-value of the sharp absorption line is about 2.02, it is reasonable to conclude that the sharp absorption line observed above TN is the EPR. Then what is the origin of the sharp absorption line below TN? The possibility of the impurity resonance can be excluded because it has the comparable intensity with EPR in Fig. 1 while almost no resonance was observed below TN in the X-band ESR measurement [4]. In order to clarify the origin of both broad and sharp absorption lines, we performed the frequency dependence measurements at 1.8 K. A typical result is shown in Fig. 3 for H//c*. The g-value of the sharp absorption line shows almost no frequency dependence while that of broad absorption line increases as the frequency is decreased. The frequency dependences for...
H//a* and H//b* show similar tendency. In the antiferromagnetic resonance (AFMR) with an uniaxial anisotropy, the g-values increase as the frequency is decreased when the field is applied along the hard axes while it decreases when the field is applied along the easy axis [7]. It turned out that both broad and sharp absorption lines do not show the frequency dependence of AFMR.

High field ESR measurements of Me₄As[Pd(dmit)₂]₂ have been performed. Although EPR was identified, the origin of the two absorption lines observed below T_N remains open for future investigation.

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References