Pressure-induced anomalous electronic phases of $\lambda\text{-}(BEDSe\text{-}TTF)_2GaCl_4$ probed by $^{69,71}Ga~NMR$

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 λ - D_2 GaCl₄ (D = BETS, BEDT-STF, ET, BEDSe-TTF) are bandwidth-controlled organic conductors by donor molecular substitution. The electronic system changes from superconducting (SC) phase to antiferromagnetic (AF) phase via no ordered phase as the bandwidth decreases [1,2]. This electronic phase transition is different from that in κ -type ET and TMTSF salts, where AF phase is adjacent to SC phase [3,4]. Further clarification of the universal phase diagram of λ - D_2 GaCl₄ is required for a comprehensive understanding of the electron-correlation effect. The proposed no ordered phase is based on experimental results for λ -(BEDT-STF)₂GaCl₄ [2]; however, it is disputable whether the no ordered phase is intrinsic or not since the disorder originating from the asymmetry of the BEDT-STF molecule may affect the electronic system. To establish the universal phase diagram of λ - D_2 GaCl₄, we need to study the change in electronic states by applying pressure to antiferromagnets without disorder. In this study, we performed ^{69,71}Ga-NMR measurements under pressure on an AF Mott insulator λ -(BEDSe-TTF)₂GaCl₄ (hereafter λ -BEDSe).

Figure 1 shows the temperature dependence of ⁷¹Ga-NMR spin-lattice relaxation rate divided by temperature, $1/T_1T$, of λ -BEDSe. At ambient pressure (a. p.), we observed a divergence of $1/T_1T$ towards $T_N = 22$ K and spectral broadening below T_N due to the AF transition, as observed in ¹³C NMR [5]. As pressure is applied, T_N decreases, and eventually at 1.0 GPa, $1/T_1T$ decreases in proportion to $T^{3/2}$ without significant spectral broadening down to 6 K. At 2.5 GPa, $1/T_1T$ peaks at 40 K with the line broadening, indicating that magnetic ordering is induced again. Namely, there is a no ordered phase between the magnetically ordered phases, although λ -BEDSe at 1.0 GPa is still on the negative pressure side of λ -(BEDT-STF)₂GaCl₄

according to our preliminary results of electrical resistivity measurements. Thus, we found that several electronic phases exist between the AF and SC phases in the universal phase diagram of λ -type salts, in contrast to the previously proposed one. In this presentation, more detailed pressure variation and the results of ¹³C NMR under pressure will be presented.

References

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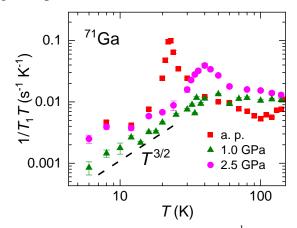


Figure 1 Temperature dependence of $(T_1T)^{-1}$ at a. p., 1.0 GPa, and 2.5 GPa in the low temperature region.