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Electronic States in Magnetic Fullerides Studied by ESR under Pressure

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Abstract

TDAE-C_{60} and AC_{60} are well known monoanion-C_{60} compounds, where TDAE is tetrakis-di-methylaminoethylene and A is Rb or Cs. The former compound is the soft ferromagnet (FM) below 16 K and the latter is a [2+2]-cycloadduct 1D polymer with the antiferromagnetic (AF) ground state below 50 K. Although several mechanisms for the FM in TDAE-C_{60} have been proposed, the present study under high pressures suggests the ordering of orbitals of Jahn-Teller distorted C_{60}. So far, AF transition below 50 K in RbC_{60} has been interpreted as the metal-inslator transition driven by instability of half-filled 1D Fermi surface. However, the present study combined with the resistivity data under pressure revealed RbC_{60} to be a 3D Mott-Hubbard system with the AF-metallic phase arising from magnetic frustration in a fcc-like structure.

Keywords: Fullerides, ESR, Magnetic phase transition, Pressure, Orbital order, polymer

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1. Introduction

To obtain new information on mechanisms dominating the phase transitions of the mono-anion fullerides, hydrostatic pressure is applied, which modifies the interaction strengths between electrons, and electron and lattice. Powder (RbC_{60}) and single crystal (TDAE-C_{60}) for ESR are located in a clamp-type pressure cell with Daphne 7373 oil. Pressure decrease at low temperatures was corrected with reported data [1]. ESR frequencies are between several tens and several hundreds MHz. Magnetic phase transition temperatures are determined from sudden changes in the relative magnetic susceptibility deduced by ESR and NMR intensities of the sample and cell. Main results and analyses have already been reported [2-4].

2. Results and Discussion

Figure 1 shows a phase diagram for orthorhombic (o-) RbC_{60} deduced from the spin susceptibility [2] togegher with the electrical resistivity under pressure [5]. With increasing pressure, AF ground state is suppressed to disappear around 12 kbar. A remarkable feature is a presence of antiferromagnetic-metal phase, typical of magnetically frustrating Mott-Hubbard systems [6], such as a fundamental fcc-like structure of o-RbC_{60}. A border of a Metal-Insulator transition $P_{M\text{-}I}$ locates around 1-2 kbar at 50 K, which is much lower than 6-8 kbar at 300 K [5]. These facts are consistent with the Mott-Hubbard system that is dominated by a transfer

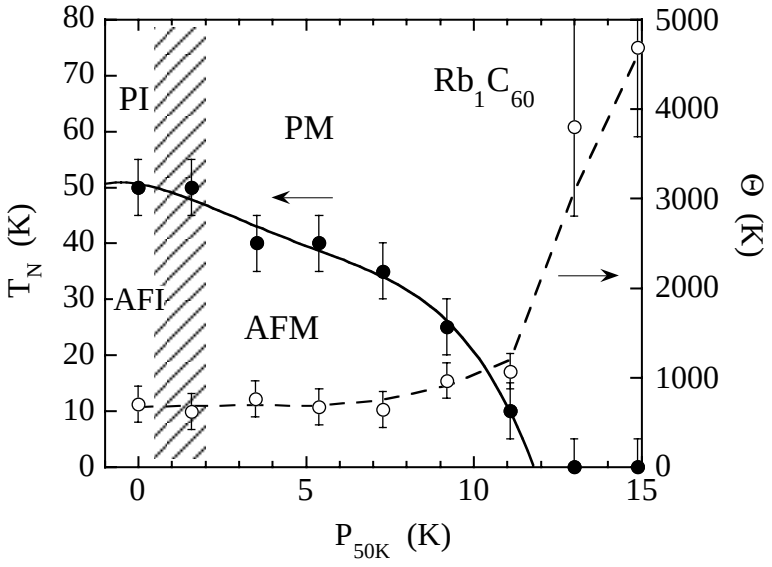

Fig. 1. *T* vs *P* diagram in o-RbC_{60}. PI: Paramagnetic Insulator, PM: Paramagnetic Metal, AFI: Antiferromagnetic Insulator and AFM: Antiferromagnetic Metal.

energy t_{inter} between C_{60} balls in the neighboring chains. The decrease of $P_{M\text{-}I}$ with decreasing temperature would be accounted for by sizable thermal contraction of the lattice constants between the chains, which dominates t_{inter}. An interpretation of AFI ground state with a nesting gap of 1D metallic Fermi-surface concluded experimentally, so far, contradicts the presence of both PI and AFM phases.

Figure 2 shows the pressure dependence of the ferromagnetic transition temperature, T_C in the other mono-anion fulleride, TDAE-C_{60} [3]. The solid curve indicates a theoretical prediction based on a two-band Hubbard model of collectively Jahn-Teller distorted C_{60} balls, where the ferromagnetic interaction is dominant with an alternating (antiferro-) ordering of the Jahn-Teller orbitals, but the interaction becomes antiferromagnetic with a homogeneous (ferro-) ordering [4]. It is noted that the first study under the pressure reported a completely different result [7], attributable to a nature of the powder sample [3]. A new phase of TDAE-C_{60}, β-phase is found; a [2+2] cycloadduct polymerization along the c-axis [3]. Above 10 kbar, the ferromagnetic α-phase irreversibly transforms to the paramagnetic β-phase that is still stable at ambient pressure up to 430 K [8]. The Curie susceptibility and the larger chain spacing of β-phase than o-RbC_{60} suggests that a single [2+2] cycloadduct polymer of C_{60}^- is insulating.

3. Conclusion

Finally, we conclude as follows on the mono-anion fullerides, RbC_{60} and TDAE-C_{60}:

1. Electronic states of RbC_{60} at ambient is near the border of a 3D Mott-Hubbard transition, instead of SDW caused by 1D-instability of the metallic Fermi-surface,

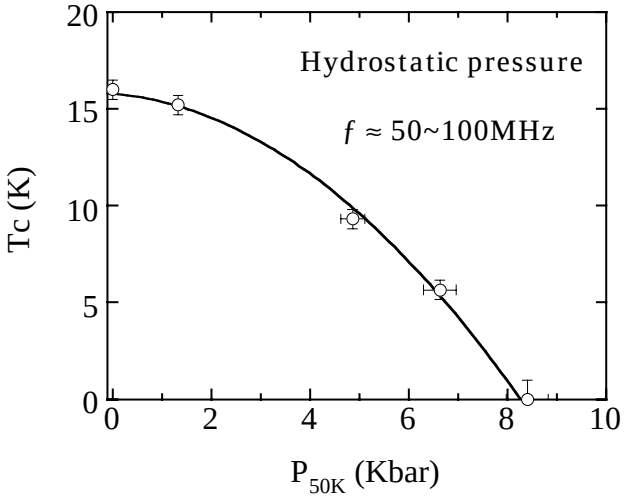

Fig. 2. The pressure dependence of the Curie temperature in α-TDAE-C_{60}.

2. Orbital ordering mechanism reproduces $T_C(P)$ in TDAE-C_{60},

3. Pressure-induced irreversible [2+2] polymer phase (β-phase) was found in TDAE-C_{60},

4. Single [2+2] polymer chain of mono-anion C_{60}'s is possibly 1D Mott-insulator.

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4. References

[1] K. Murata, H. Yoshino, H. O. Yadav, Y. Honda, and N. Shirakawa, Rev. Sci. Instrum. 68, (1997) 2490.

[2] H. Sakamoto, S. Kobayashi, K. Mizoguchi, M. Kosaka and K. Tanigaki, Phys. Rev. B 62, (2000) R7691.

[3] K. Mizoguchi, M. Machino, H. Sakamoto, T. Kawamoto, M. Tokumoto, A. Omerzu, and D. Mihailovic, Phys. Rev. B 63, (2001) 140417(R).

[4] T. Kawamoto, M. Tokumoto, H. Sakamoto, and K. Mizoguchi, J. Phys. Soc. Jpn. 70, (2001) 1892.

[5] K. Khazeni, V. H. Crespi, J. Hone, A. Zettl and M. L. Cohen, Phys. Rev. B 56, (1997) 6627.

[6] G. Kotliar and G. Moeller, in: A. Fujimori, Y. Tokura (Eds)., Springer Series in Solid-State Sciences, vol. 119, Springer, Tokyo, 1994, pp 15-27.

[7] G. Sparn, J. D. Thompson, P.-M. Allemand, Q. Li, F. Wudl, K. Holczer and P. W. Stephens, Solid St. Commun. 82, (1992) 779.

[8] K. Mizoguchi, M. Machino, H. Sakamoto, T. Kawamoto, M. Tokumoto, A. Omerzu, and D. Mihailovic, in: S. Saito (Ed)., Proceedings of ISNM2001 (AIP, Kamakura, 2001).