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Spin soliton dynamics and pressure effects in the spin-Peierls system $(DMe\text{-}DCNQI)_2M$ (M = Li, Ag)

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Abstract

$(DMe\text{-}DCNQI)_2M$ (M = Li, Ag) is a $\frac{1}{4}$-filled spin-Peierls system. We study the spin/charge dynamics in the insulating state by EPR. The linewidth shows exponential dependence at $T \gg T_{sp}$. Further, pressure enhances mainly prefactor of the exponential dependences. These results are successfully understood in terms of the relaxation accompanied with the intercolumn hopping caused by the spin–orbit interaction both for the Li and Ag salts. This is consistent with the scenario that the hole and spin solitons help the intercolumn charge and spin transports.

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Keywords: ESR; Spin-Peierls; High pressure; Soliton dynamics

Since the $\frac{1}{4}$-filled electronic systems, $(DMe\text{-}DCNQI)_2Li$ and $(DMe\text{-}DCNQI)_2Ag$ are isostructural, they show similar physical properties; the spin-Peierls (SP) transition at ≈ 65 K for the Li salt and ≈ 80 K for the Ag salt [1,2]. Above T_{SP}, $4k_F$ charge density wave (CDW) gap with dimerization in the half-filled lower Hubbard band caused by large on-site Coulomb repulsion U dominates the electronic properties. Since the charges localize because of U, these systems are expected to have a peculiar charge transport mechanism which leads to the relatively high conductivity $\sigma \approx 10$–100 S/cm [1,2]. Here, we report on the spin/charge dynamics in these salts studied by EPR.

A single crystal is used for W-band (94 GHz) EPR but an assembly of the single crystals is utilized for 50 MHz EPR under pressure. A clamp-type pressure cell made of Cu–Be alloy is used with Daphne 7373 pressure medium.

Fig. 1 shows the hydrostatic pressure dependence of T_{SP} up to 1.8 GPa. The monotonic increase of T_{SP} suggests one dimensionality (1D) of this system preserving up to 1.8 GPa. This fact is consistent with the structure of this material; a network of the DCNQI columns isolated by the metal ions preserving 1D nature even under pressure, and enhanced overlap of the π-electrons in the DCNQI column which could enhance T_{SP} [3,4].

In Fig. 2 the EPR linewidth under several pressures are shown for the Li salt, together with the Ag salt in the inset. As demonstrated with the solid curves for the Li salt, the temperature dependence of the linewidth is well reproduced by an empirical exponential dependence as reported earlier [2]. Here, note that the pressure does not modify the activation energy of the formula, $T_{Li} = 250$ K, but mainly changes the prefactor of the exponential formula. Interpretation of this point will be given after identification of the mechanism of the EPR linewidth.

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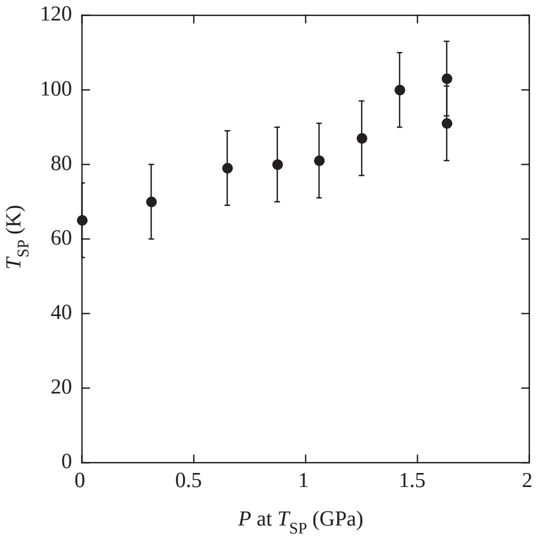

Fig. 1. The pressure dependence of T_{SP} for the Li salt.

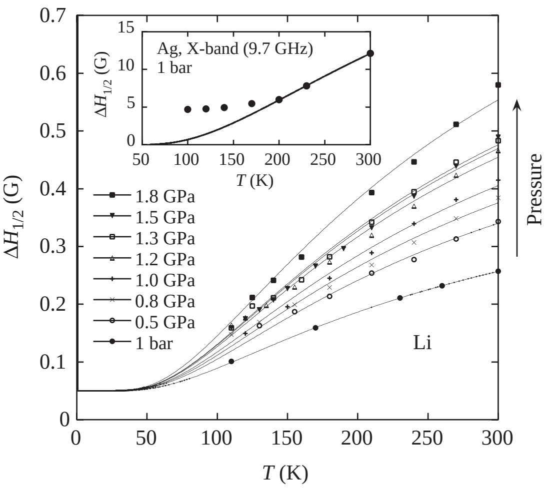

Fig. 2. Temperature dependence of ESR linewidth taken at 50 MHz in the powder Li salt above T_{SP} with implicit parameter of hydrostatic pressure up to 1.8 GPa. The inset shows the linewidth for Ag salt at X-band and ambient pressure. Solid lines represent an empirical formula, $0.05 + A(P)\exp(-250/T)$ for the Li salt and $B(P)\exp(-400/T)$ for the Ag salt, where $A(0) = 0.5$ G and $B(0) = 50$ G.

First, we examine the electron–electron dipolar interaction with the strong exchange interaction J as an origin of the linewidth [5]. The estimated second moment of 70 G with $J \approx 500$ K deduced from the spin susceptibility gives the EPR linewidth of ≈ 1 mG, fairly smaller than the observed value of 0.23 G for the Li salt at 300 K.

A hint is found in the temperature dependence of the intercolumn hopping rate $D_\perp$ reproduced by the same formula as the linewidth determined with angular dependence of the EPR linewidth at W-band; $D_\perp = 1 \times 10^{10} \exp(-250/T)$ for the Li salt and $2 \times 10^{11} \exp(-400/T)$ for the Ag salt. The electron spin can be flipped if the spin is allowed to hop to the neighboring column with a help of the spin–orbit interaction [6]. In such a case, the transition probability is proportional to the square of g-shift. With $\Delta g = 0.0011$ for the Li salt and 0.0025 for the Ag salt, the ratio of the expected linewidth for the Ag salt with respect to the linewidth of the Li salt is obtained as $103 \exp(-400/T)/\exp(-250/T)$ which agrees very well with the same ratio for the observed linewidth, $100 \exp(-400/T)/\exp(-250/T)$. To validate this mechanism, vacant DCNQI site is inevitable to avoid the loss of U, supporting a scenario of the pair creation of hole and spin solitons with fractional charge $\pm e/2$ thermally excited in the $4k_F$-CDW gap. Based on this conclusion, the limited effect of pressure to the exponential prefactor in the linewidth is ascribed to the enhancement of the overlap between DCNQI and Ag wavefunctions which enhances $D_\perp$. Further details will be published elsewhere.

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