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# Quasi-one dimensional diffusive motion of spin solitons in the spin-Peierls state of $(DMe\text{-}DCNQI)_2Li$

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**Abstract**

$(DMe\text{-}DCNQI)_2Li$ is a 1/4 filled spin-Peierls (SP) system with the SP transition temperature $T_{sp} \sim 65$K. Using frequency variable ESR at temperatures enough below $T_{sp}$, the existance of spin solitons which could arise from domains with finite chain length, is demonstrated. It is also found that $T_{sp}$ is enhanced by hydrostatic pressure and can be reproduced well with Cross-Fisher's relation. Within our knowledge, this is the first example in such good agreement with theory in pressure dependence of $T_{sp}$. It is due to a simple quasi-1-D electronic structure of $(DMe\text{-}DCNQI)_2Li$, even under high pressures. Such a simple agreement with the SP theoretical prediction would be a unique feature in this system.

*Key words:* spin-Peierls; ESR; soliton dynamics; high pressure

## 1. Introduction

Except for Cu salt, DCNQI electronic systems are dominated by a strong Coulomb correlation and are highly one dimensional, which gives rise to various low dimensional electronic phenomena. In a sense that Li has no *d* electrons, $(DMe\text{-}DCNQI)_2Li$ is the most fundamental compound among DCNQI salts. However, there still remain a lot of questions about electronic states, such as a charge transfer mechanism in a higher temperature range. As a recent progress, the presence of the spin solitons and their spin dynamics were unveiled [1].

## 2. Experimental

Paramagnetic spin susceptibilities and quasi one dimensional (Q1D) spin dynamics of $(DMe\text{-}DCNQI)_2Li$ were measured using ESR under hydrostatic pressure. The spin susceptibility was calibrated with the $^{19}F$-NMR intensity in a sample tube. The spin dynamics were investigated with relaxation rate at various frequencies between 10MHz and 35GHz. The frequency dependence of the ESR line width (spin-spin relaxation) reflects anisotropic spin motion.

## 3. Results and Discussion

The spin susceptibility at 1GPa is shown in Fig. 1. At ambient pressure and 1.5GPa, it also shows a similar tendency to that at 1GPa. The $T_{sp}$ derived from the susceptibility is indicated in Fig. 2. From Cross-Fisher's calculation [2], $T_{sp}$ is expressed by $0.8\lambda^2 J^2/Ka^2$, where $\lambda$ is the spin-phonon coupling parameter, $J$ the nearest neighbor spin exchange interaction, $K$ the elastic constant, and $a$ the lattice constant. Now we estimate the pressure effect on $T_{sp}$. In a Mott-Hubbard insulator with $U >> t$, where $U$ is the on-site Coulomb interaction, $J$ depends on a square of transfer integral $t$. Since the wavefunction is strongly

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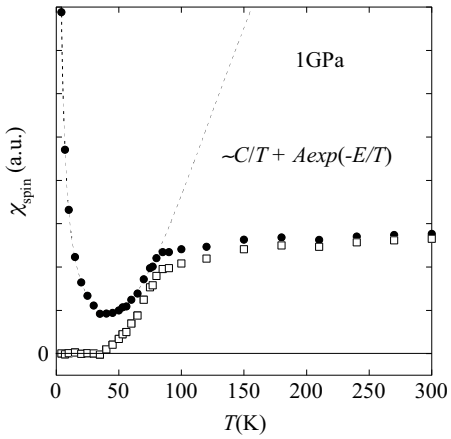


Fig. 1. The electronic spin susceptibility at 1GPa obtained with ESR (50MHz). Closed circles show the raw data. Open circles show that obtained by subtracting Curie contributions from the raw data. They disappear exponentially below $T_{sp}$.

localized, an overlap of wavefunctions dominating $t$ is small. Then, $t$ depends on the overlap of exponential tails of the wavefunctions. Since $J$ is modified through the lattice contraction caused by pressure, $T_{sp}$ also varies with pressure. That is, the transfer integral is given by $t = t_0 \exp[-(a - a_0)/r_0]$, where $a_0$ is the lattice constant, $t_0$ the transfer integral at ambient pressure, and $r_0$ the characteristic decay length of the wavefunction. And $J \propto t^2 \propto \exp[-2(a - a_0)/r_0]$. Substituting $J$ for the $T_{sp}$ formula, with an expansion of $a(P)$ to first order in $P$; $a(P) \sim a_0 + \partial a/\partial P \cdot P$, we obtain the first approximation of the pressure dependence of $T_{sp}(P)$, as

$$T_{sp}(P) = 0.8\frac{\lambda^2 J^2(P)}{Ka^2} \propto e^{-4\frac{\partial a'}{\partial P}\frac{a_0}{r_0}P}, \qquad (1)$$

where $a'$ is the normalized lattice constant $a' = a/a_0$. Here, the relatively smaller pressure variations than $J^2$ are neglected for the other parameters in Eq. (1). From Fig. 2, the compressibility $\partial a'/\partial P$ is obtained as $0.075 r_0/a_0$ ($\mathrm{GPa}^{-1}$). Since the typical compressibility of Van der Waals bonding is $\sim 0.02$ ($\mathrm{GPa}^{-1}$), $r_0 \sim a_0/4$ is expected, which is consistent with the localized wavefunctions in the Mott-Hubbard insulator. Then, $(\mathrm{DMe\text{-}DCNQI})_2\mathrm{Li}$ looks like an ideal 1-D SP system so that the theory can reproduce the pressure dependence of $T_{sp}$.

Figure 3 shows the Q1D spin dynamics between 4.2K and 300K. The intra- and inter- chain diffusion rates $D_{//}$ and $D_\perp$ were obtained from the frequency dependence of ESR line width [1] under ambient pressure, 1GPa, and 1.5GPa. All the results are quite similar to each other at temperatures enough below $T_{sp}$. In this temperature range, $D_{//} \sim T^2$, which is similar to that of the neutral solitons in $t$-$(\mathrm{CH})_x$ [3]. Near the $T_{sp}$, $D_{//}$ begins to increase rapidly. We will discuss $D_\perp$ in future publications with the higher accuracy data.

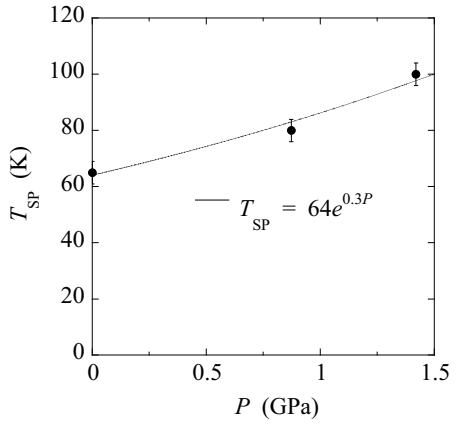


Fig. 2. The pressure dependence of $T_{SP}$. The solid curve represents Eq. (1).

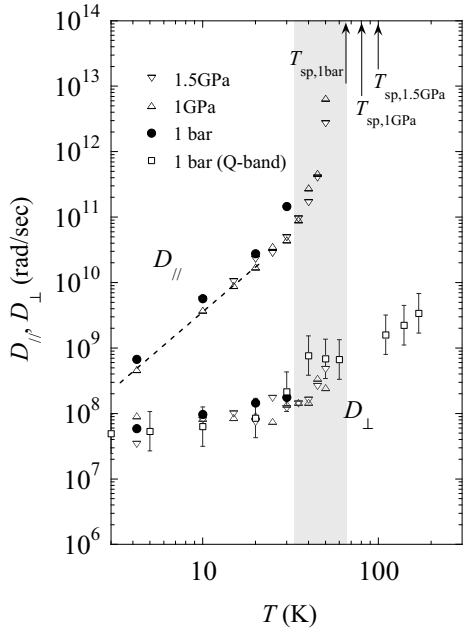


Fig. 3. The temperature dependence of $D_{//}$ and $D_\perp$. Crosses, open triangles, and closed circles are deduced from the frequency dependence of ESR line width (10MHz∼2GHz) [1]. Open squares are estimated from the motional broadening at Q-band (35GHz) ESR. We will discuss their details in future publication.

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