



## NMR relaxation rate and dynamical structure factors in nematic and multipolar liquids of frustrated spin chains under magnetic fields

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Recently, it has been shown that spin nematic (quadrupolar) or higher multipolar correlation functions exhibit a quasi-long-range order in the wide region of the field-induced Tomonaga-Luttinger-liquid (TLL) phase in spin- $\frac{1}{2}$  zigzag chains. In this Rapid Communication, we point out that the temperature dependence of the NMR relaxation rate  $1/T_1$  in these multipolar TLLs is qualitatively different from that in more conventional TLLs of one-dimensional quantum magnets (e.g., the spin- $\frac{1}{2}$  Heisenberg chain);  $1/T_1$  decreases with lowering temperature in multipolar TLL. We also discuss low-energy features in spin dynamical structure factors which are characteristic of the multipolar TLL phases.

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Magnetic states with an order parameter defined by a product of multiple spins, such as nematic, vector chiral, and scalar chiral orders, have attracted much attention. Spin nematic-(quadrupolar)-ordered phases have been recently shown to appear in frustrated ferromagnets such as ferromagnets with competing antiferromagnetic (AF) interactions<sup>1-3</sup> and magnets with multi-spin-exchange couplings.<sup>2,4,5</sup> A triatic-(octupolar)-ordered phase was also found in the triangular lattice multiple-spin-exchange model with ferromagnetic (FM) dominant coupling.<sup>3</sup> These nematic- and triatic-ordered states can be regarded as Bose-condensed states of bound two magnons<sup>1,2</sup> and bound three magnons,<sup>3</sup> respectively, and their order parameters are given by  $S_j^+ S_k^+$  or  $S_j^- S_k^-$  and  $S_j^+ S_k^+ S_l^+$  or  $S_j^- S_k^- S_l^-$ .

Recent extensive studies<sup>6-10</sup> have shown that a series of similar multipolar phases appear in the one-dimensional (1D) spin- $\frac{1}{2}$  Heisenberg model with FM nearest-neighbor exchange  $J_1$  and competing AF next-nearest-neighbor exchange  $J_2$  in applied field  $H$ , whose Hamiltonian is

$$\mathcal{H} = \sum_{n=1,2} \sum_j J_n \mathbf{S}_j \cdot \mathbf{S}_{j+n} - H \sum_j S_j^z. \quad (1)$$

Here  $S_j$  is the spin- $\frac{1}{2}$  operator on  $j$ th site,  $J_1 < 0$  and  $J_2 > 0$ , and  $H$  is the external magnetic field in the  $z$  direction. This simple frustrated spin chain is a minimal model of frustrated ferromagnets and is thought to describe magnetism in quasi-one-dimensional (quasi-1D) edge-sharing cuprates<sup>11-14</sup> such as  $\text{Rb}_2\text{Cu}_2\text{Mo}_3\text{O}_{12}$ ,  $\text{NaCu}_2\text{O}_2$ ,  $\text{LiCuVO}_4$ , and  $\text{LiCu}_2\text{O}_2$ .

Hikihara *et al.*<sup>9</sup> and Sudan *et al.*<sup>10</sup> showed that the ground state of Hamiltonian (1) has field-induced Tomonaga-Luttinger-(TL)-liquid phases in which spin multipolar correlations are quasi-long-range ordered while the transverse spin correlation is short ranged. This result can be easily understood in the large-magnetization regime, where  $p$  magnons form a bound state ( $p > 1$ ).<sup>8,9</sup> A gas of bound  $p$  magnons acquires off-diagonal quasi-long-range order, with the order parameter being the effective hard-core boson creation operator  $\prod_{l=1, \dots, p} S_{j+l}^-$ . In the original spin language, this off-diagonal correlation is the multipolar spin correlation

characterizing a nematic ( $p=2$ ), an octupolar ( $p=3$ ), or a hexadecapolar ( $p=4$ ) phase. Numerical studies<sup>8-10</sup> found  $p=2$  for  $-2.7 \leq J_1/J_2 < 0$ ,  $p=3$  for  $-3.5 \leq J_1/J_2 \leq -2.7$ , and  $p=4$  for  $-3.76 \leq J_1/J_2 \leq -3.5$  at the saturation field. At lower magnetic fields these phases cross over to spin-density wave  $\text{SDW}_p$  phases in which the density correlation of bound  $p$  magnons, i.e., the longitudinal SDW, becomes stronger than the multipolar correlation. Incidentally, a  $\text{SDW}_2$  phase is present also in the case of AF  $J_1 > 0$ .<sup>15,16</sup>

However, it will be difficult to obtain direct experimental evidence for the multipolar spin orders, as it requires probing four—or more—spin-correlation functions with high accuracy. Standard experimental probes, such as neutron scattering or magnetic resonance, measure only two-spin correlations. Furthermore, the multipolar TL liquids have a gapless spectrum and a smooth magnetization curve. Thus, if one only measures their static thermodynamic quantities (uniform susceptibility, specific heat, entropy, etc.), it is hard to distinguish the multipolar TL liquids from conventional TL liquids. Experimental schemes for identifying multipolar spin orders are therefore called for.

In this Rapid Communication, we propose that NMR measurements can capture signatures (albeit indirect) of the multipolar TL liquids. We show that in the TL liquids with a dominant multipolar spin correlation, the NMR relaxation rate  $1/T_1$  decreases as temperature  $T$  is lowered [see Fig. 1(a)]. This temperature dependence of  $1/T_1$  is opposite to that in conventional TL liquids (and in  $\text{SDW}_p$  phases), where it is always diverging as  $T \rightarrow 0$ , in magnetic field.<sup>17</sup> We also point out that the spin dynamical structure factors exhibit features which are very characteristic of the multipolar TL liquids.

Let us begin with a brief review of the effective theories<sup>7,9</sup> for the multipolar TL liquids, which allow us to find low-energy behavior of spin-correlation functions observed in NMR and in dynamical structure factors. In the weak  $J_1$  limit, the Abelian bosonization method<sup>18</sup> is useful. It leads to nematic ( $p=2$ ) and  $\text{SDW}_2$  phases<sup>6,7,9</sup> and a vector chiral-ordered phase.<sup>19</sup> The effective Hamiltonian for the nematic ( $p=2$ ) and  $\text{SDW}_2$  phases<sup>6,7,9</sup> is written as

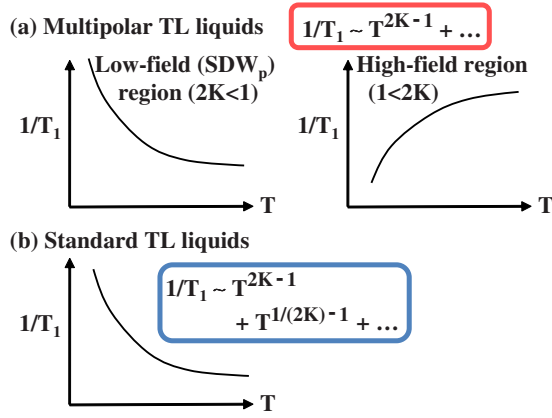


FIG. 1. (Color online) Temperature and field dependence of the NMR relaxation rate  $1/T_1$  in (a) multipolar phases in spin- $\frac{1}{2}$ -frustrated zigzag chains and in (b) standard TL liquids (e.g., the AF Heisenberg chain in magnetic field). The parameter  $K$  controls correlations in TL liquids (see the text).

$$\mathcal{H}_{\text{eff}} = \int dx \left\{ \sum_{\nu=\pm} \frac{v_{\nu}}{2} [K_{\nu}^{-1} (\partial_x \phi_{\nu})^2 + K_{\nu} (\partial_x \theta_{\nu})^2] + g \sin(\pi M) \sin(\sqrt{8\pi} \phi_{-} + \pi M) \right\}, \quad (2)$$

where  $x=2j$  (the lattice spacing is set equal to unity),  $(\phi_{\pm}, \theta_{\pm})$  is a pair of dual scalar fields satisfying the commutator  $[\phi_{\mu}(x), \partial_y \theta_{\nu}(y)] = i \delta_{\mu,\nu} \delta(x-y)$ ,  $M = \langle S_j^z \rangle$ ,  $g \propto J_1$ , and  $K_{\pm}$  and  $v_{\pm}$  are, respectively, the TL-liquid parameter and the velocity of the  $(\phi_{\pm}, \theta_{\pm})$  sector. The  $(\phi_{+}, \theta_{+})$  sector is a gapless TL liquid, while the  $(\phi_{-}, \theta_{-})$  sector has a gapful spectrum because the field  $\phi_{-}$  is pinned at a value minimizing the potential energy of the  $g$  term. Using Hamiltonian (2), one can evaluate the low-energy and long-distance behaviors of several correlation functions. The imaginary-time ( $\tau$ ) spin and nematic correlations at zero temperature  $T=0$  are calculated as

$$\langle S_j^z(\tau) S_0^z(0) \rangle = M^2 - \frac{K_{+}}{2\pi^2} \left( \frac{1}{z_{+}^2} + \frac{1}{z_{-}^2} \right) + \frac{C_1}{|z_{+}|^{K_{+}}} \cos \left[ \pi j \left( M + \frac{\text{sgn}(J_1)}{2} \right) \right] + \dots, \quad (3a)$$

$$\langle S_j^+(\tau) S_0^-(0) \rangle = C_2 \cos \left( \frac{\pi j}{2} \right) \frac{e^{-|z_{-}|/\xi}}{|z_{+}|^{1/(4K_{+})} |z_{-}|^{1/2}} + \dots, \quad (3b)$$

$$\langle S_j^+(\tau) S_{j+1}^+(\tau) S_0^-(0) S_1^-(0) \rangle = C_3 \frac{(-1)^j}{|z_{+}|^{1/K_{+}}} + \dots, \quad (3c)$$

where  $z_{\pm} = j - iv_{\pm} \tau$ , and  $C_n$  are nonuniversal positive constants. The exponential decay of the transverse spin correlation in Eq. (3) is qualitatively different from a power-law decay form in ordinary TL liquids (e.g., the spin- $\frac{1}{2}$  AF chain and ladder in magnetic field). The correlation length  $\xi$  is inversely proportional to the gap of the  $(\phi_{-}, \theta_{-})$  sector.

More generally, the TL-liquid behavior in *all* the multipolar and SDW $_p$  phases ( $p \geq 2$ ) can be understood from a hard-core Bose gas picture of bound  $p$  magnons, when the nearest-neighbor coupling  $J_1$  is ferromagnetic.<sup>8,9</sup> Below the saturation field, a (dilute) Bose gas of bound  $p$  magnons forms a TL liquid with off-diagonal quasi-long-range order, i.e.,  $p$ th multipolar TL liquid. In this picture, one may replace the  $p$ th multipolar operator  $S_{j+1}^- S_{j+2}^- \cdots S_{j+p}^-$  and magnon density  $\frac{1}{2} - S_j^z$  with a creation operator of a hard-core boson  $(-1)^j b_j^\dagger$  and boson density  $p b_j^\dagger b_j$ , respectively. Here the staggered factor  $(-1)^j$  represents the total momentum  $k = \pi$  of the lowest-energy bound states. The hydrodynamic theory for the bosonic TL liquid has the same form as the free boson Hamiltonian of the  $(\phi_{+}, \theta_{+})$  sector in Eq. (2). The effective theory gives the following longitudinal spin and the multipolar correlation functions at  $T=0$ :

$$\langle S_j^z(\tau) S_0^z(0) \rangle = M^2 - \frac{p^2 K}{4\pi^2} \left( \frac{1}{z^2} + \frac{1}{z^2} \right) + \frac{C_4 p^2}{|z|^{2K}} \cos \left[ \frac{\pi j}{p} (1 - 2M) \right] + \dots, \quad (4a)$$

$$\left\langle \prod_{n=1}^p S_{j+n}^+(\tau) \prod_{n=1}^p S_n^-(0) \right\rangle = C_5 \frac{(-1)^j}{|z|^{1/(2K)}} + \dots, \quad (4b)$$

where  $z = j - iv\tau$ ,  $K$  is the TL-liquid parameter for the hard-core bosons. While we cannot evaluate the transverse spin correlations within this boson picture, they must decay exponentially  $\propto \exp(-|z|/\xi)$  as it is necessary to break a magnon bound state in order to create an excitation with  $\Delta S^z = \pm 1$ . In the nematic case of  $p=2$  and  $J_1 < 0$ , Eq. (4) coincides with Eqs. (3a) and (3c) if we set  $K_{+} = 2K$  and  $v_{+} = v$ . Near the saturation field where the density of magnons vanishes, the value of  $K$  approaches unity, i.e., that of the 1D free fermions. Indeed, the numerical calculations in Ref. 9 have shown that  $K$  monotonically increases from about 1/4 to unity with the increase in the magnetization  $M$ . This means that the multipolar correlation (4b) is strongest in the high-field regime ( $2K > 1$ ), whereas the SDW correlation (4a) becomes most dominant in the low-field regime ( $2K < 1$ ). This property is important in the following discussion on the NMR relaxation rate. We note that at  $p=1$ , Eq. (4) reproduces the spin correlations in the TL-liquid phase of, e.g., the spin- $\frac{1}{2}$  AF chains under magnetic field.<sup>18</sup>

The temperature dependence of the NMR relaxation rate  $1/T_1$  in the multipolar TL liquids can be derived from the above asymptotic forms of correlation functions. The perturbation theory in hyperfine interaction between nuclear and electron spins obtains  $1/T_1$  as<sup>17,20</sup>

$$\frac{1}{T_1} \propto \sum_k \left\{ \frac{|A_k^\perp|^2}{2} [S^{+-}(k, \omega) + S^{-+}(k, \omega)] + |A_k^\parallel|^2 S^{zz}(k, \omega) \right\}, \quad (5)$$

where  $\omega$  is the nuclear resonance frequency,  $A_k^\nu$  are the hyperfine form factors, and  $S^{\alpha\beta}(k, \omega) = \sum_j e^{-ikj} \int_{-\infty}^{\infty} dt e^{i\omega t} \langle S_j^\alpha(t) S_0^\beta(0) \rangle$  is the spin dynamical structure factor ( $t = -i\tau$  is the real time) at temperature  $T$ . Since  $\omega$  is

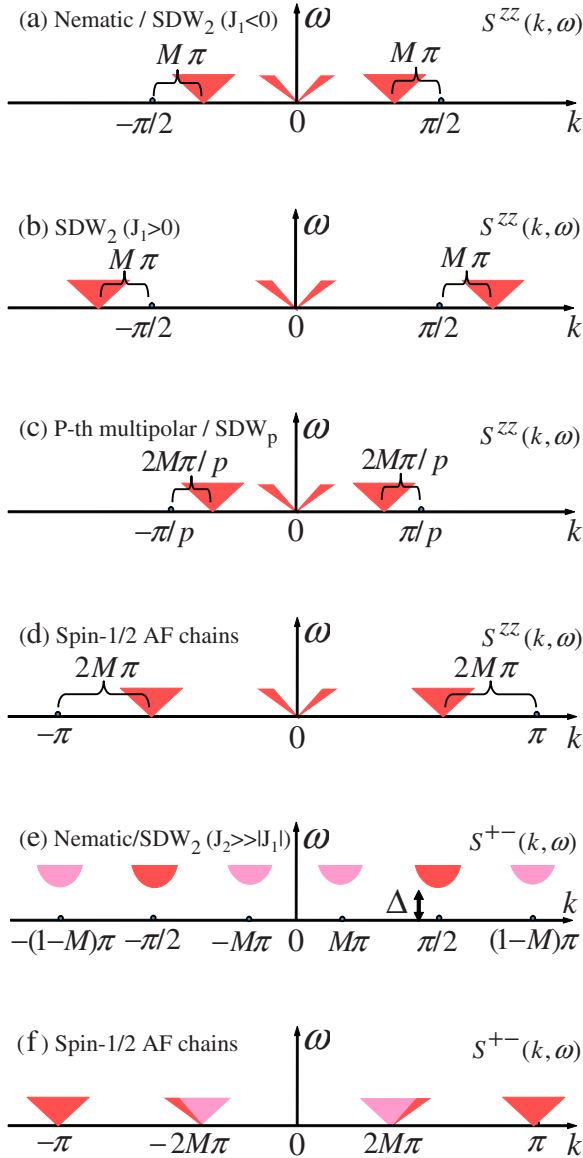


FIG. 2. (Color online) Low-energy relevant parts of  $S^{zz}(k, \omega)$  and  $S^{+-}(k, \omega)$  in the multipolar and  $\text{SDW}_p$  phases of the spin- $\frac{1}{2}$ -frustrated zigzag chain in magnetic field [(a)–(c) and (e)] and in the TL liquid of spin- $\frac{1}{2}$  AF chains in magnetic field [(d) and (f)].

generally much smaller than the energy scale of spin-exchange interactions, we may take the limit  $\omega/T \rightarrow +0$ . Moreover, the  $k$  dependence of  $A_k^p$  is usually weak due to the locality of the nucleus-electron interaction. Hence, the  $T$  dependence of  $1/T_1$  can be obtained by evaluating the local susceptibility  $\int_{-\infty}^{\infty} dt e^{i\omega t} \langle S_j^{\alpha}(t) S_j^{\beta}(0) \rangle$ .

The local susceptibility at finite temperatures can be readily obtained from the correlation functions (3) and (4) through the standard procedure.<sup>17,18</sup> Substituting them into Eq. (5), we obtain  $1/T_1$  for the multipolar TL liquids in the form

$$1/T_1 = D_1^{\parallel} T + D_2^{\parallel} T^{2K-1} + \dots \quad (6)$$

The two leading terms come from the second and third terms, respectively, of the longitudinal spin correlation (3) or

(4). The coefficients  $D_1^{\parallel}$  and  $D_2^{\parallel}$  are independent of temperature in the regime  $\omega \ll T \ll |J_{1,2}|$ . In Eq. (6) we have omitted contributions from the transverse spin correlations which are exponentially small,  $e^{-\Delta/T}$  ( $\Delta \approx v/\xi$  is proportional to the spin gap) at low temperatures  $T \ll |J_{1,2}|$ . When  $K < 1$ , the second term in Eq. (6) gives the leading contribution in the low-temperature limit. Similarly, the known  $T$  dependence of  $1/T_1$  in spin- $\frac{1}{2}$  AF chains under a magnetic field is obtained from Eq. (3) with  $p=1$  in the form<sup>17,18</sup>

$$1/T_1 = E_1^{\parallel} T + E_2^{\parallel} T^{2K-1} + E_1^{\perp} T^{1/(2K)-1} + \dots, \quad (7)$$

where the terms  $\propto E_n^{\parallel}$  and  $E_n^{\perp}$  are derived from the longitudinal and the transverse spin correlations, respectively. Equation (7) commonly holds in TL-liquid phases of 1D magnets such as AF spin chains and ladders in magnetic field.<sup>17</sup>

Comparison of Eqs. (6) and (7) tells us an important feature of the NMR relaxation rate in the multipolar TL liquids. As we noted above, the parameter  $K$  in the multipolar phases of Hamiltonian (1) with the FM coupling  $J_1 < 0$  is an increasing function of  $H$  and approaches unity at the saturation field.<sup>9</sup> The monotonic magnetic-field dependence of  $K$  presumably holds for other multipolar TL liquids as well, at least for spin- $\frac{1}{2}$  AF spin systems. Equation (6) then implies that  $1/T_1$  decreases with lowering temperature in the high-field multipolar phase ( $2K > 1$ ),<sup>21</sup> while it shows diverging behavior in the low-field  $\text{SDW}_p$  region ( $2K < 1$ ) (see Fig. 1). This behavior is totally different from that of conventional TL liquids, such as AF spin chains under magnetic field [Eq. (7)], in which  $1/T_1$  always diverges in the low-temperature limit irrespective of the value of  $K$  (the case of  $K=1/2$  is special<sup>22,23</sup>). We emphasize that this difference in the  $T$  dependence of  $1/T_1$  between multipolar and conventional TL liquids shown in Figs. 1(a) and 1(b) can be taken as a pronounced signature of 1D spin- $\frac{1}{2}$  multipolar TL-liquid phases. The decay of  $1/T_1$  with lowering temperature in the multipolar liquid phases is due to both the absence of gapless modes in  $S^{+-}(k, \omega)$  and the weak singularity at  $\omega=0$  in  $S^{zz}(k, \omega)$  [see Eqs. (8) and (9)]. We also note that NMR experiments cannot distinguish a  $\text{SDW}_p$  region from ordinary TL liquids because they both show divergent behavior of  $1/T_1$  as  $T \rightarrow 0$ .

Next we discuss the spin dynamical structure factors  $S^{\alpha\beta}(k, \omega)$  at  $T=0$  in the multipolar phases. The support of  $S^{\alpha\beta}(k, \omega)$  tells us which excitations in the  $(k, \omega)$  space contribute to inelastic neutron scattering. The low-energy parts of  $S^{\alpha\beta}(k, \omega)$  are obtained from Fourier transform<sup>24</sup> of correlation functions (3) and (4). For the nematic and  $\text{SDW}_2$  phases in FM  $J_1 < 0$ , we find

$$S^{zz}(k \sim 0, \omega) = 4K|k| \delta(\omega - v|k|), \quad (8a)$$

$$S^{zz}(k \sim \pm k_2, \omega) = \frac{c_2^z \Theta_s(\omega - v|k \mp k_2|)}{[\omega^2 + v^2(k \mp k_2)^2]^{1-K}}, \quad (8b)$$

$$S^{+-}(k \sim \pm \pi/2, \omega) = \frac{c_2^{\pm} \Theta_s[\omega - \epsilon(k \mp \pi/2)]}{[\omega - \epsilon(k \mp \pi/2)]^{1-1/(4K_+)}}, \quad (8c)$$

where  $k_2 = \pi(1-2M)/2$ ,  $\epsilon(k) = (v^2 k^2 + \Delta^2)^{1/2}$ ,  $\Theta_s(\omega)$  is a unit step function, and  $c_p^{\pm}$  are positive numerical constants. The  $\delta$ -function peak in Eq. (8) will have a finite width when the

nonlinearity of the low-energy dispersion is included. In the SDW<sub>2</sub> phase with AF  $J_1 > 0$ ,  $k_2$  in Eq. (8) should be replaced with  $\tilde{k}_2 = \pi(1+2M)/2$ . The longitudinal part  $S^{zz}(k, \omega)$  in the higher multipolar and SDW <sub>$p$</sub>  phases ( $p \geq 2$ ) is also obtained from Eq. (4) as

$$S^{zz}(k \sim 0, \omega) = p^2 K |k| \delta(\omega - v|k|), \quad (9a)$$

$$S^{zz}(k \sim \pm k_p, \omega) = \frac{c_p^z \Theta_s(\omega - v|k \mp k_p|)}{[\omega^2 + v^2(k \mp k_p)^2]^{1-K}}, \quad (9b)$$

where  $k_p = \pi(1-2M)/p$ . For comparison,  $S^{\alpha\beta}(k, \omega)$  in the standard TL liquids, e.g., spin- $\frac{1}{2}$  AF Heisenberg chains under a magnetic field, have the form<sup>18</sup>

$$S^{zz}(k \sim 0, \omega) = K |k| \delta(\omega - v|k|), \quad (10a)$$

$$S^{zz}(k \sim \pm k_1, \omega) = \frac{c_1^z \Theta_s(\omega - v|k \mp k_1|)}{[\omega^2 + v^2(k \mp k_1)^2]^{1-K}}, \quad (10b)$$

$$S^{+-}(k \sim \pm \pi, \omega) = \frac{c_1^{\pm} \Theta_s(\omega - v|k \mp \pi|)}{[\omega^2 + v^2(k \mp \pi)^2]^{1-1/(4K)}}, \quad (10c)$$

$$S^{+-}(k \sim \pm 2\pi M, \omega) = \frac{\tilde{c}_1^{\pm} \Theta_s(\omega - v|k \mp 2\pi M|)}{[\omega \mp v(k \mp 2\pi M)]^2} \\ \times [\omega^2 - v^2(k \mp 2\pi M)^2]^\gamma, \quad (10d)$$

where  $\gamma = K + \frac{1}{4K}$ , and  $\tilde{c}_1^{\pm}$  is a positive constant.

These results are depicted in Fig. 2. The gapless excitations giving dominant contribution to  $S^{zz}(k, \omega)$  in the  $p$ th multipolar or the SDW <sub>$p$</sub>  TL liquid are located at  $k = \pm(1-2M)\pi/p$ , when  $J_1 < 0$ . These wave numbers in-

versely proportional to the number  $p$  of magnons forming a bound state are equal to the “ $2k_F$ ” of the hard-core Bose liquid of bound  $p$  magnons<sup>9</sup> (note that fermions and hard-core bosons are equivalent in one dimension). The result for the ordinary TL liquid (e.g., the AF Heisenberg chain) corresponds to the case  $p=1$  or the limit  $J_1 \rightarrow 0$  (the lattice unit equals two in this case). Furthermore, one can discriminate between the SDW<sub>2</sub> phases in  $J_1 < 0$  and in  $J_1 > 0$  by observing the shift of the gapless points from  $k = \pi/2$  in  $S^{zz}(k, \omega)$ . Another manifest difference between multipolar and ordinary TL liquids is that the transverse component  $S^{+-}(k, \omega)$  has a gap in the multipolar phases, while that of the ordinary TL liquids is gapless. These features in  $S^{\alpha\beta}(k, \omega)$  can be employed as definite signatures of the multipolar and the SDW <sub>$p$</sub>  phases in the model (1).

To conclude, we have studied dynamical response of the multipolar TL liquids in the spin- $\frac{1}{2}$ -frustrated zigzag chains in applied magnetic field. The NMR relaxation rate  $1/T_1$  in the multipolar TL liquids shows algebraic decay with lowering temperature, which is distinct from the diverging behavior in conventional TL liquids like the spin- $\frac{1}{2}$  AF chains (see Fig. 1). Furthermore, the wave number and the magnetization dependence of the gapless modes in the dynamical structure factors  $S^{\alpha\beta}(k, \omega)$  can provide us with clear evidence for the presence of the multipolar liquids as well as SDW <sub>$p$</sub>  regions of bound magnons. Our arguments are also applicable to multipolar phases in higher-spin chains, where  $K$  can become larger than unity due to soft-core repulsion of bosons.

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