Field and temperature dependence of NMR relaxation rate in the magnetic quadrupolar liquid phase of spin-$\frac{1}{2}$ frustrated ferromagnetic chains

Masahiro Sato,[1,2] Toshiya Hikihara,[3] and Tsutomu Momoi[1]
[1]*Condensed Matter Theory Laboratory, RIKEN, Wako, Saitama 252-5258, Japan*
[2]*Department of Physics and Mathematics, Aoyama Gakuin University, Sagamihara, Kanagawa 229-8558, Japan*
[3]*Department of Physics, Hokkaido University, Sapporo 060-0810, Japan*
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It is generally difficult experimentally to distinguish magnetic multipolar orders in spin systems. Recently, it was proposed that the temperature dependence of the nuclear magnetic resonance relaxation rate $1/T_1$ can involve an indirect but clear signature of the field-induced spin nematic or multipolar Tomonaga-Luttinger (TL) liquid phase [Phys. Rev. B **79**, 060406(R) (2009)]. In this paper, we evaluate accurately the field and temperature dependence of $1/T_1$ in spin-$\frac{1}{2}$ frustrated J_1-J_2 chains combining field-theoretical techniques with numerical data. Our results demonstrate that isotherms of $1/T_1$ as a function of magnetic field also exhibit distinctive nonmonotonic behavior in spin nematic TL liquid, in contrast with the standard TL liquid in the spin-$\frac{1}{2}$ Heisenberg chain. The relevance of our results to quasi-one-dimensional edge-sharing cuprate magnets, such as $LiCuVO_4$, is discussed.

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I. INTRODUCTION

One of the current topics in solid-state magnetism is multiple-spin ordering without any single-spin dipole moment.[1,2] Vector and scalar chiral orders and spin multipolar orders are typical examples expected to appear in real compounds. The emergence of vector and scalar chiralities accompanies the spontaneous breakdown of parity or time-reversal symmetries, and these two could be detected indirectly from parity- or time-reversal-odd observables and related quantities (e.g., electric polarization in multiferroics, asymmetric momentum dependence of spin structure factors, Hall conductivity, etc.). On the other hand, spin multipolar orders, for example, spin nematic and spin triatic orders, in spin-$\frac{1}{2}$ magnets can occur without breaking spatial symmetry, since they are characterized as condensation of bound multimagnons[3–8] or a spin-triplet resonating valence bond state.[9] The absence of both spin long-range order and lattice symmetry breaking makes it difficult to find them in experiments. Furthermore, no clear experimental proof of the spin multipolar orders has ever been reported, because there is no established experimental method of probing spin multipolar orders.

Thanks to recent theoretical studies, it has been gradually recognized that magnetic multipolar states occur in several geometrically frustrated spin-$\frac{1}{2}$ magnets, especially in low dimensions.[3–8,10–14] In this paper, we focus on spin-$\frac{1}{2}$ frustrated chains with ferromagnetic (FM) nearest-neighboring exchange $J_1 < 0$ and competing antiferromagnetic (AF) next-nearest-neighboring exchange $J_2 > 0$. The Hamiltonian is given by

$$\mathcal{H}_{J_1-J_2} = \sum_{n=1,2} \sum_j J_n \boldsymbol{S}_j \cdot \boldsymbol{S}_{j+n} - H \sum_j S_j^z, \qquad (1)$$

where $\boldsymbol{S}_j$ is the spin-$\frac{1}{2}$ operator on the jth site and H is the applied magnetic field along the z axis. Recent studies revealed that this model possesses a series of field-induced multipolar Tomonaga-Luttinger (TL) liquid phases in high magnetization regime.[7,8,10,11,15] These multipolar TL liquids are interpreted as a hard-core Bose gas of multimagnon bound states,[7,8] where the number p of magnons forming a multimagnon bound state changes successively from $p = 2$ to 4 with varying $J_2/|J_1|$. In the multipolar phase with $p = 2$, the quadrupolar (or spin nematic) operator $S_j^{\pm} S_{j+1}^{\pm}$ exhibits a quasi-long-range order. Similarly, the octupolar (or triatic) operator $S_j^{\pm} S_{j+1}^{\pm} S_{j+2}^{\pm}$ and the hexadecapolar operator $S_j^{\pm} S_{j+1}^{\pm} S_{j+2}^{\pm} S_{j+3}^{\pm}$ show a quasi-long-range order, respectively, in the phases with $p = 3$ and 4. In all of these phases, the longitudinal spin correlation also decays algebraically, while the transverse spin correlation is short-ranged. These characteristic properties of the multipolar TL liquid phases are distinct from those in the standard TL liquid (e.g., spin-$\frac{1}{2}$ Heisenberg chain). Numerical studies[8,11] of the quadrupolar and octupolar TL liquid phases showed that the relevant multipolar correlations are dominant in the high-magnetization regime, whereas the longitudinal spin-density-wave (SDW) correlation becomes dominant in the lower-magnetization regime. We call the high-field multipolar states with $p = 2$ and 3 *quadrupolar and octupolar liquids* and the low-field multipolar states *SDW_2 and SDW_3 liquids*, respectively, following Ref. 8.

In these multipolar TL liquids, bulk static quantities, such as entropy, specific heat, and uniform susceptibility, exhibit qualitatively the same behaviors as those of standard TL liquids, and hence it is impossible to distinguish multipolar and standard TL liquids from them. To detect direct evidence of their ordering, we need to measure multiple-spin order parameter or relevant multiple-spin correlation functions, which is generally difficult. Instead, to find any indirect evidence, it is necessary to specify an effective method of detecting any signature of spin multipolar ordering through usual experiments. Recently, Ref. 15 has proposed an experimental way of detecting a signature of multipolar TL liquids, showing that the nuclear magnetic resonance (NMR) relaxation rate $1/T_1$ decreases with lowering temperature T in the multipolar TL liquids near saturation. This is completely different from the behavior of standard TL liquids in one-dimensional (1D) AF magnets such as the spin-$\frac{1}{2}$ Heisenberg chain, in which $1/T_1$ always increases with lowering T irrespective of the value of applied magnetic field H. However, it is generally hard to experimentally

approach the vicinity of the saturation field, especially in magnets with strong exchange couplings. In fact, the saturation field of a J_1-J_2 magnet $LiCuVO_4$ is about $40-50$ T,[16,17] and to perform NMR measurement under such a high field is not an easy task. An experimentally detecting scheme at low magnetic field is therefore desirable. It was also shown in Ref. 15 that the momentum dependence of dynamical structure factors in the multipolar TL liquids shows a distinct difference from that of the spin-$\frac{1}{2}$ AF Heisenberg chain.

In this paper, we reexamine the NMR relaxation rate $1/T_1$ in the spin quadrupolar (nematic) and SDW_2 TL liquids, including both low- and high-magnetic-field regimes, and evaluate its field and temperature dependence quantitatively. To this end, we combine the field-theoretical (bosonization) approach with the density-matrix renormalization-group (DMRG) method, substituting numerical values for nonuniversal parameters in analytic results. We compare these results with the relaxation rate $1/T_1$ of the spin-$\frac{1}{2}$ AF Heisenberg chain. It is found that, in addition to the temperature dependence, the field dependence of $1/T_1$ in the nematic and SDW_2 liquids clearly differs from that in the usual TL liquid of the spin-$\frac{1}{2}$ AF chain. In the J_1-J_2 chain, the relaxation rate $1/T_1$ slowly decreases with an increase of magnetic field (or is almost independent of the field) in the low-field SDW_2 regime and increases suddenly in the high-field nematic regime near saturation, whereas it increases monotonically, as a function of the field, in the usual TL liquid of the spin-$\frac{1}{2}$ AF chain. The nonmonotonic field dependence of $1/T_1$ is a unique characteristic of the multipolar TL liquids in spin-$\frac{1}{2}$ J_1-J_2 chains. Furthermore, if we tune the direction of the field H, we can eliminate the dominant contribution to $1/T_1$ in the multipolar phases and thereby make $1/T_1$ show exponential decays at low temperature in both nematic and SDW_2 liquids. This exponential decay is caused by a spin gap in the spin transverse excitations, which is also a unique property of the multipolar TL liquids. Our prediction would be applicable in NMR experiments of quasi-1D edge-sharing cuprate magnets, such as $LiCuVO_4$ (Refs. 16–19), $Rb_2Cu_2Mo_3O_{12}$ (Ref. 20), $PbCuSO_4(OH)_2$ (Refs. 21–24), $LiCu_2O_2$ (Refs. 25–28), and $NaCu_2O_2$ (Ref. 29), whose magnetic properties are expected to be described by the spin-$\frac{1}{2}$ J_1-J_2 model with FM J_1 and AF J_2.

The paper is organized as follows. In Secs. II and III, we review briefly the theory of the NMR relaxation rate $1/T_1$ in electron-spin systems and effective theories for multipolar and usual TL liquids. Particularly, in Sec. III, relying on an established field-theoretical method, we write down the formula of $1/T_1$ in multipolar liquid phases of the J_1-J_2 chain (1) and also that in TL liquid of the spin-$\frac{1}{2}$ AF Heisenberg chain. Section IV contains the main results of the present paper, which are quantitative estimates of $1/T_1$ in both quadrupolar and usual TL liquids as a function of magnetic field and temperature. Section V is devoted to a discussion of some relevant factors in real magnets, which are neglected in the previous sections. We consider a broad temperature scale beyond the low-energy effective theory, the relation between electron-nuclear spin interaction and direction of field H, and the effects of Dzyaloshinsky-Moriya interaction. Tuning of the field direction is also discussed. Finally, in Sec. VI, we summarize our results and their relevance to real quasi-1D compounds.

II. NMR RELAXATION RATE

Here, we briefly explain the formula of the NMR relaxation rate $1/T_1$ in electron-spin systems. Provided that the NMR relaxation process is mainly caused by interaction between electron spin $\boldsymbol{S}_j$ and nuclear spin $\boldsymbol{I}$, the standard perturbation theory for the interaction evaluates the relaxation rate $1/T_1$ as follows:[30,31]

$$1/T_1 \propto \sum_k \frac{1}{2}|\tilde{A}_\perp(k)|^2[\mathcal{S}^{+-}(k,\omega)+\mathcal{S}^{-+}(k,\omega)] + |\tilde{A}_\parallel(k)|^2\mathcal{S}^{zz}(k,\omega). \tag{2}$$

Here, we have assumed that the electron-spin system is in one dimension. The electron-spin dynamical structure factor at finite temperature T is defined as

$$\mathcal{S}^{\mu\nu}(k,\omega)=\sum_j e^{-ikj}\int_{-\infty}^{\infty} dt\ e^{i\omega t}\left\langle S_j^\mu(t)S_0^\nu(0)\right\rangle_T, \tag{3}$$

where $\langle\cdots\rangle_T$ denotes the thermal average. The frequency ω in Eq. (2) is a given resonant value of applied oscillating field. Since its magnitude is generally much smaller than the energy scale of electron systems, we may take a limit $\omega/k_BT=\beta\omega\to 0$ in Eq. (2). The symbols $\tilde{A}_\perp(k)$ and $\tilde{A}_\parallel(k)$ denote Fourier components of hyperfine coupling constants between electron and nuclear spins, which generally stem from the dipole-dipole interaction and an SU(2)-invariant exchange interaction $\boldsymbol{S}_j\cdot\boldsymbol{I}$. The longitudinal component $\tilde{A}_\parallel(k)$ originates only from the dipole-dipole interaction. In quantum spin systems, $\tilde{A}_\parallel(k)$ is usually the same order as $\tilde{A}_\perp(k)$.

The spatial range of interactions between electron spins and a single nuclear spin is local, that is, at most the order of the lattice spacing a. Therefore, the k dependence of $\tilde{A}_{\perp,\parallel}(k)$ could be negligible. Under the assumption of such a locality, the hyperfine coupling term in the Hamiltonian is given by

$$\mathcal{H}_{\rm hf}=S_{j=0}^\mu\mathcal{A}_{\mu\nu}I^\nu, \tag{4}$$

where the electron site closest to the nucleus is assumed to be $j=0$ and $\mathcal{A}_{\mu\nu}$ is the real-space hyperfine coupling tensor. In this case, $1/T_1$ can be approximated as

$$1/T_1 \propto \tfrac{1}{2}A_\perp^2(j=0)[\mathcal{S}_{j=0}^{+-}(\omega)+\mathcal{S}_{j=0}^{-+}(\omega)] + A_\parallel^2(j=0)\mathcal{S}_{j=0}^{zz}(\omega). \tag{5}$$

Here, hyperfine couplings $A_\perp(0)$ and $A_\parallel(0)$ are proportional to $\mathcal{A}_{xx}+\mathcal{A}_{yy}$ and $(\mathcal{A}_{xz}^2+\mathcal{A}_{yz}^2)^{1/2}$, respectively. The local dynamical structure factor $\mathcal{S}_{j=0}^{\mu\nu}(\omega)$ at finite temperature is represented as

$$\mathcal{S}_{j=0}^{\mu\nu}(\omega)=\int_{-\infty}^{\infty} dt\ e^{i\omega t}\left\langle S_0^\mu(t)S_0^\nu(0)\right\rangle_T. \tag{6}$$

III. MULTIPOLAR AND USUAL TL LIQUIDS

In this section, we briefly review the effective theory for multipolar TL-liquid phases in the J_1-J_2 spin chain (1) and the

well-established TL-liquid theory in the spin-$\frac{1}{2}$ AF Heisenberg chain. Using these theories, we can derive analytic forms of dynamical structure factors and $1/T_1$.

A. Theory for multipolar liquid phases

Two analytic approaches have been developed for field-induced multipolar phases in the model (1): One is a weak-coupling approach[8,10] where $|J_1|/J_2 \ll 1$ is assumed and J_1 is treated as a perturbation for two decoupled AF-J_2 chains. The other is a phenomenological bosonization in the vicinity of the saturation field.[7,8] The former weak-coupling theory can lead to only the quadrupolar (nematic) phase, but it enables us to calculate various physical quantities in principle. The latter approach can treat all of the multipolar phases (quadrupolar, octupolar, and hexadecapolar), while the quantities that can be evaluated are somewhat restricted. Here, we employ the latter approach, which is sufficient to calculate $1/T_1$ (the former will be used in Sec. V).

In the J_1-J_2 chain with ferromagnetic $J_1 < 0$, the excitation mode which destabilizes the fully polarized state at the saturation field H_c is a multimagnon bound state consisting of $p(\geqslant 2)$ magnons. Below the saturation field, the soft bound magnons proliferate to form TL liquid with dominant transverse multipolar correlation. The system is thus described as a hard-core Bose gas of p-magnon bound states. From a numerical calculation of the p-magnon excitation energies, it has been shown that the soft mode to realize the multipolar TL liquid is two-, three-, and four-magnon bound states with momentum $k = \pi$ for $-2.669 < J_1/J_2 < 0$, $-3.514 < J_1/J_2 < -2.720$, and $-3.764 < J_1/J_2 < -3.514$, respectively.[7,32]

From the picture of the hard-core Bose gas, we may map multipolar operators and the z component of spin operators to operators of bosons in the following forms:

$$\begin{aligned} b_j^\dagger &= (-1)^j S_j^- \cdots S_{j+p-1}^-, \\ n_j \equiv b_j^\dagger b_j &= \frac{1}{p}\left(\frac{1}{2} - S_j^z\right), \end{aligned} \tag{7}$$

where b_j is an annihilation operator of the hard-core boson, and we have considered the pth-order multipolar liquid states ($p = 2, 3$, and 4 correspond to quadrupolar, octupolar, and hexadecapolar liquids, respectively). The staggered factor $(-1)^j$ in the first line comes from the momentum $k = \pi$ of the soft multimagnon bound states. To the 1D hard-core Bose gas, we can apply the standard Abelian bosonization.[33,34] Its low-energy effective Hamiltonian is represented as

$$\mathcal{H}^{\rm eff}_{J_1-J_2} = \int dx \frac{u}{2}[\kappa^{-1}(\partial_x \Phi)^2 + \kappa(\partial_x \Theta)^2], \tag{8}$$

where $x = ja$ is a spatial coordinate, Φ and Θ are a canonical pair of scalar fields, κ is the TL-liquid parameter, and u is the elementary excitation velocity. We set $\kappa \to 1$ here at the saturation limit $H \to H_c$ where the hard-core boson gas becomes equal to a free fermion gas. It is numerically shown[8,11] that the TL-liquid parameter κ decreases almost monotonically with lowering magnetic field H in quadrupolar and octupolar liquid phases. The velocity u is expected to be of the order of $|J_{1,2}|a$, except for the saturation limit $H \to H_c$, where $u \to 0$. We note that the TL-liquid theory is valid only in the temperature regime of $k_B T \ll u/a$.

The boson operator b_j and the density operator n_j are expressed in terms of Φ and Θ:[33,34]

$$\begin{aligned} b_j^\dagger &\approx e^{i\sqrt{\pi}\Theta}[f_0 + f_1 \cos(\sqrt{4\pi}\Phi + 2\pi\rho j) + \cdots], \\ n_j &\approx \rho + \frac{a}{\sqrt{\pi}}\partial_x \Phi + g_0 \cos(\sqrt{4\pi}\Phi + 2\pi\rho j) + \cdots, \end{aligned} \tag{9}$$

where $\rho = (1/2 - M)/p$ is the mean value of the boson density and $M = \langle S_j^z \rangle$ is the magnetization per site. Factors f_n and g_n are nonuniversal constants. In the present representation, the scaling dimensions of $e^{qi\sqrt{4\pi}\Phi}$ and $e^{qi\sqrt{\pi}\Theta}$ are, respectively, $q^2\kappa$ and $q^2/(4\kappa)$.

From Eqs. (7)–(9), two-point correlation functions of S_j^z and the pth multipolar operator $M_j^{(p)} = \prod_{n=1}^p S_{j+n-1}^-$ at zero temperature are shown to behave asymptotically like

$$\begin{aligned} \left\langle S_j^z(\tau) S_0^z(0)\right\rangle_0 &\approx M^2 - \frac{p^2\kappa}{4\pi^2}\left[\left(\frac{a}{w}\right)^2 + \left(\frac{a}{\bar{w}}\right)^2\right] \\ &\quad + \frac{g_0^2}{2}\cos(2\pi\rho j)\left(\frac{a}{|w|}\right)^{2\kappa} + \cdots, \\ \left\langle M_j^{(p)}(\tau) M_0^{(p)\dagger}(0)\right\rangle_0 &\approx (-1)^j f_0^2 \left(\frac{a}{|w|}\right)^{1/(2\kappa)} + \cdots \end{aligned} \tag{10}$$

in the pth multipolar TL-liquid phase, where $\langle \cdots \rangle_0$ denotes the expectation value of observables at zero temperature. (Incidentally, the $p = 1$ case corresponds to the usual TL liquid in the spin-$\frac{1}{2}$ AF chain. See the following subsection.) Here, $\tau = it$ is imaginary time, and light-cone coordinates w and $\bar{w}$ are defined as $(w, \bar{w}) = (x + iu\tau, x - iu\tau)$. Equation (10) has been numerically confirmed,[8,10,11] and this asymptotic behavior survives down to the phase boundary between multipolar and lower-field vector chiral phases. The power-law behavior also indicates that in the high-field regime (narrowly defined multipolar regime) with $\kappa > 1/2$, the multipolar correlation is stronger than the longitudinal spin correlation, while the latter is dominant in the lower-field SDW regime with $\kappa < 1/2$. Namely, $\kappa = 1/2$ is the crossover point between multipolar and SDW regimes.

In the multipolar TL liquid phase, the transverse spin correlation decays exponentially, since a finite energy is necessary to violate p-magnon bound states. Though we cannot compute the transverse spin correlation function and the multipolar correlation functions $\langle M_j^{(p')}(\tau) M_0^{(p')\dagger}(0)\rangle_0$ with $p' < p$ within the theory of hard-core Bose gas of multimagnon bound states, we can explicitly derive the form of the transverse spin correlation function $\langle S_j^{\pm}(\tau) S_0^{\mp}(0)\rangle_0$, which is short-ranged,[15] using the weak-coupling theory in the quadrupolar-liquid phase ($p = 2$). Furthermore, the exponential decay of the p' ($<p$) magnon correlations including the transverse spin correlation has been confirmed numerically for the entire regime of the quadrupolar and octupolar TL liquid phases.[8] The single-magnon gap can be determined by measuring transverse dynamical structure factors[15] $\mathcal{S}^{\pm\mp}(k,\omega)$, for example, in neutron scattering experiment. It would be at most the order of $|J_1|$ or J_2. This fact of the short-ranged transverse spin correlation in the multipolar TL liquids plays a key role in the following argument.

The slowly decaying mode in the longitudinal spin correlator in Eq. (10) contributes to the leading temperature dependence of NMR relaxation rate $1/T_1$. Through an established method based on conformal field theory,[34–38] we obtain the local dynamical structure factors at finite temperature from correlation functions at zero temperature and achieve the formula for $1/T_1$,

$$1/T_1 \propto A_{\parallel:0}^2(0)\frac{p^2\kappa}{\pi}\left(\frac{a}{u}\right)^2\beta^{-1} + A_{\parallel:1}^2(0)\frac{g_0^2}{2}\frac{2a}{u} \times \cos(\kappa\pi)B(\kappa,1-2\kappa)\left(\frac{2\pi a}{\beta u}\right)^{2\kappa-1} + \cdots, \quad (11)$$

where $B(x,y)$ is the beta function and $A_{\parallel:0}(j) = A_{\parallel:1}(j) = A_{\parallel}(j)$. The first and second terms on the right-hand side are, respectively, contributions from the second and third terms of $\langle S_j^z(\tau)S_0^z(0)\rangle_0$ in Eq. (10). If we take into account the spatial dependence of $A_{\parallel}(j)$ instead of the formula (5), $A_{\parallel:n}(j)$ generally take different values (but they are expected to be the same order). The transverse spin correlation also contributes to $1/T_1$, but it must be a thermal activation type $\sim e^{-\Delta/(k_BT)}$, in which Δ is proportional to the binding energy of p magnons, that is, the single-magnon gap. Therefore, it can be negligible when temperature is sufficiently smaller than Δ/k_B. The absence of the contribution from the transverse spin correlation is a characteristic feature of the multipolar TL liquids.

The temperature dependence of $1/T_1$ given in Eq. (11) is consistent with the previous prediction in Ref. 15. The NMR relaxation rate $1/T_1$ diverges with lowering temperature in the SDW regime ($\kappa < 1/2$), whereas it decays algebraically with lowering T in the high-field multipolar regime ($\kappa > 1/2$). Thus the temperature dependence changes significantly at the crossover point $\kappa = 1/2$. This feature contrasts sharply with the result of usual TL liquids (see the next subsection). More detailed properties of $1/T_1$ will be discussed in Sec. IV.

B. TL-liquid theory for the spin-$\frac{1}{2}$ AF chain

Here, we quickly summarize the well-known effective theory for the spin-$\frac{1}{2}$ AF Heisenberg chain in magnetic field to compare with the previously mentioned theory for multipolar liquids. The Hamiltonian is given by

$$\mathcal{H}_J = \sum_j J\boldsymbol{S}_j\cdot\boldsymbol{S}_{j+1} - H\sum_j S_j^z, \quad (12)$$

where $J > 0$ is AF exchange coupling. As is well known, the low-energy physics of this model belongs to the TL-liquid universality class from zero field to the saturation field $H_c = 2J$. The low-energy Hamiltonian has the same free-boson form as Eq. (8):

$$\mathcal{H}_J^{\text{eff}} = \int dx\frac{v}{2}[K^{-1}(\partial_x\phi)^2 + K(\partial_x\theta)^2], \quad (13)$$

where ϕ and θ are the pair of scalar fields. The TL-liquid parameter K and the velocity v are exactly determined from the Bethe ansatz.[39–42] In this model, K changes monotonically from $1/2$ to 1 when H increases from zero to H_c. Similarly to the multipolar TL liquids, the value of v is of order of Ja except for the saturation limit $H \to H_c$, where $v \to 0$. The TL-liquid theory is valid only for $k_BT \ll v/a$.

Spin operators are bosonized as

$$S_j^z \approx M + \frac{a}{\sqrt{\pi}}\partial_x\phi + (-1)^j a_1\cos(\sqrt{4\pi}\phi + 2\pi Mj) + \cdots, \quad (14)$$
$$S_j^+ \approx e^{i\sqrt{\pi}\theta}[b_0(-1)^j + b_1\cos(\sqrt{4\pi}\phi + 2\pi Mj) + \cdots].$$

Here a_1, b_0, and b_1 are nonuniversal coefficients, whose values are numerically determined as a function of H (or M) in Ref. 43. Equations (13) and (14) enable us to estimate $1/T_1$. The result is

$$1/T_1 \propto A_{\parallel:0}^2(0)\frac{K}{\pi}\left(\frac{a}{v}\right)^2\beta^{-1} + A_{\parallel:1}^2(0)\frac{a_1^2}{2}\frac{2a}{v}\cos(K\pi)B(K,1-2K)\left(\frac{2\pi a}{\beta v}\right)^{2K-1} + A_{\perp:1}^2(0)b_0^2\frac{2a}{v}\cos\left(\frac{\pi}{4K}\right)B\left(\frac{1}{4K},1-\frac{1}{2K}\right) \times \left(\frac{2\pi a}{\beta v}\right)^{1/(2K)-1} + \cdots, \quad (15)$$

where $A_{\parallel:0}(j) = A_{\parallel:1}(j) = A_{\parallel}(j)$ and $A_{\perp:1}(j) = A_{\perp}(j)$. The first and second terms on the right-hand side originate from the longitudinal spin correlation, while the third term originates from the transverse spin correlation. In contrast to the multipolar liquids, both longitudinal and transverse spin correlations yield power-law-type functions of temperature. Equation (15) shows that either $A_{\parallel:1}$ or $A_{\perp:1}$ term diverges at low temperatures regardless of the value of K, except for $K = 1/2$.[44] This diverging behavior of $1/T_1$ with decreasing temperature is a common property of usual TL liquids in 1D magnets, and is quite different from the decaying behavior of $1/T_1$ in the multipolar liquid phases.

IV. FIELD AND TEMPERATURE DEPENDENCE OF $1/T_1$

Utilizing the formulas (11) and (15), we examine the magnetic-field and temperature dependence of $1/T_1$ in both multipolar and usual TL liquid phases. Among the three multipolar phases, we concentrate on the quadrupolar TL liquid, which is most widely expanded in the J_1/J_2-H space and is believed to be realized in several real magnets. We show that $1/T_1$ in the quadrupolar TL liquid exhibits characteristic features distinct from that in the usual TL liquid. In Eqs. (11) and (15), the values of hyperfine constants $A_{\parallel,\perp:n}(j)$ depend on crystal structure, the spatial relationship between electron and nuclear spins, and the direction of applied field H. For simplicity, we assume that all the constants $A_{\parallel,\perp:n}(j)$ are equal throughout this section.

Let us first consider $1/T_1$ of the quadrupolar liquid phase. We can evaluate the expression of $1/T_1$ given in Eq. (11), once the parameters κ, u, and g_0 are numerically estimated. To this end, we perform numerical calculations of spin-$\frac{1}{2}$ J_1-J_2 chains using the DMRG method. For κ and g_0, we compute the equal-time longitudinal spin correlation function $\langle S_j^z(0)S_{j'}^z(0)\rangle_0$ and the spin polarization $\langle S_j^z(0)\rangle_0$ in the ground state. We then fit the data to an analytic form obtained by the bosonization method to estimate κ and g_0. (The details of the method have been presented in Refs. 8,43, and 45.) For the velocity u, we

use uniform magnetic susceptibility. From the bosonization formula (9), the susceptibility is given by

$$\chi_u = \partial\langle S_j^z\rangle/\partial H = p^2\kappa a/(\pi u) \tag{16}$$

in the pth order multipolar phase. We can thus obtain the value of u from the magnetization curve (i.e., susceptibility) determined by the DMRG calculation. We determine the values of κ, u, and g_0 at $M = 0.05, 0.1, 0.15, \ldots, 0.45$ for several values of J_1/J_2. We have found that the TL-liquid parameter κ is basically an increasing function of the field H (see Figs. 14 and 18 of Ref. 8), while g_0 has a fairly small field dependence. As expected, u/a is shown to be of order of the 1D couplings J_1 and J_2 for intermediate magnetization and approaches zero near the saturation. For example, in the case of $J_1/J_2 = -1\,(-2)$, $u/a \simeq 1.04J_2\,(0.49J_2)$ at $M = 0.25$, while $u/a \simeq 0.25J_2\,(0.1J_2)$ at $M = 0.4$.

Figure 1 shows the magnetic-field dependence of $1/T_1$ in the J_1-J_2 spin chain for $J_1/J_2 = -1$ and -2 at several values of temperature. Except for the vicinity of the saturation,

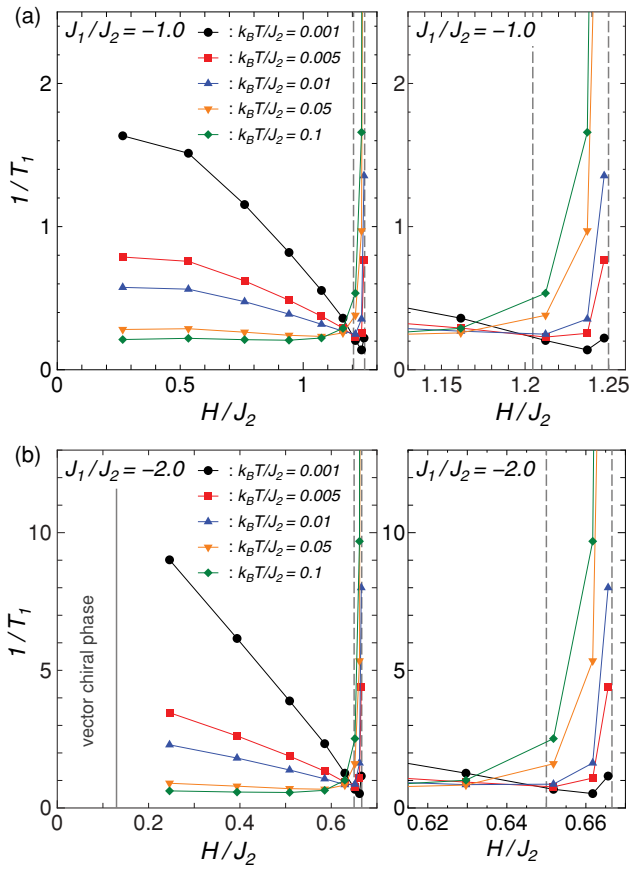

FIG. 1. (Color online) Field dependence of NMR relaxation rate $1/T_1$ in the magnetic quadrupolar TL-liquid phase in the J_1-J_2 spin chain (1) for (a) $J_1/J_2 = -1.0$ and (b) $J_1/J_2 = -2.0$. Left panels show the results for the whole range of field $0 < H < H_c$, while right panels show the same data in the vicinity of the saturation field in an enlarged scale. Solid symbols represent the data obtained from Eq. (11) with numerical values of κ, u, and g_0. Lines connecting them are guide for the eyes. The vertical dashed lines represent the saturation field and the crossover line between the SDW ($\kappa < 1/2$) and nematic ($\kappa > 1/2$) regimes. In (b), the boundary to the low-field vector-chiral phase is shown by a vertical solid line.

$1/T_1$ decreases with increasing H (or M) for sufficiently low temperatures $k_BT \ll u/a \sim |J_{1,2}|$. This is mainly due to the monotonic field dependence of the TL-liquid parameter κ, and is a distinct feature of the multipolar TL liquids, in contrast with that of the spin-$\frac{1}{2}$ AF chain (see the next paragraph). This feature becomes less significant as temperature increases, and it becomes almost independent of the field at relatively high temperatures $k_BT \sim 0.1J_2$. On the other hand, in a narrow region near the saturation field, $1/T_1$ increases rapidly with H since the velocity u approaches zero. We also confirm the previous prediction[15] on the temperature dependence of $1/T_1$: $1/T_1$ increases divergently with lowering T in the low-field SDW regime ($\kappa < 1/2$), whereas it decreases algebraically in the high-field quadrupolar regime ($\kappa > 1/2$). Explicit temperature dependence is presented in Fig. 2. The characteristic behavior in the low-temperature regime $k_BT \lesssim 0.1J_2$ is consistent with the previous statement.

Next, we compare these features of the relaxation rate in the nematic liquid with those in a usual TL liquid of the spin-$\frac{1}{2}$ AF Heisenberg chain (12). Similarly to the nematic case, we can evaluate $1/T_1$ from the formula (15) if values for the parameters K, v, a_1, and b_0 are prepared. As mentioned in Sec. III B, K and v are exactly determined by the Bethe ansatz[39–42] and the coefficients a_1 and b_0 have been evaluated numerically.[43] Using these accurate values, we numerically determine $1/T_1$ of the model (12). The result is shown in

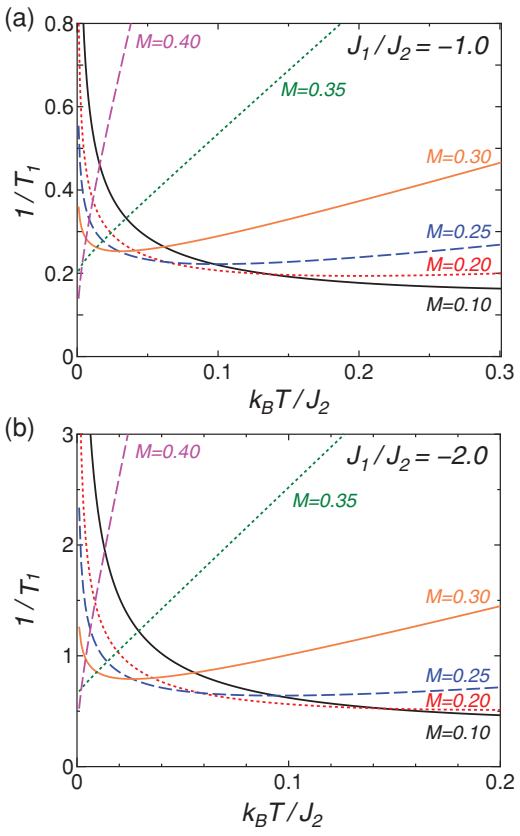

FIG. 2. (Color online) Temperature dependence of NMR relaxation rate $1/T_1$ in the magnetic quadrupolar TL-liquid phase in J_1-J_2 spin chain (1) for (a) $J_1/J_2 = -1.0$ and (b) $J_1/J_2 = -2.0$ and several fixed values of magnetization M.

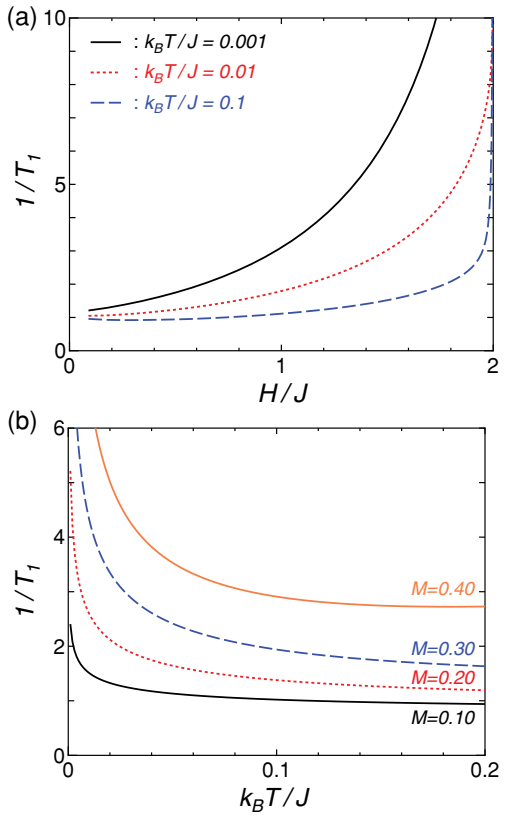

FIG. 3. (Color online) NMR relaxation rate $1/T_1$ for the standard TL-liquid phase in the spin-$\frac{1}{2}$ Heisenberg chain (12). (a) Field dependences for fixed temperatures and (b) temperature dependences for fixed magnetization.

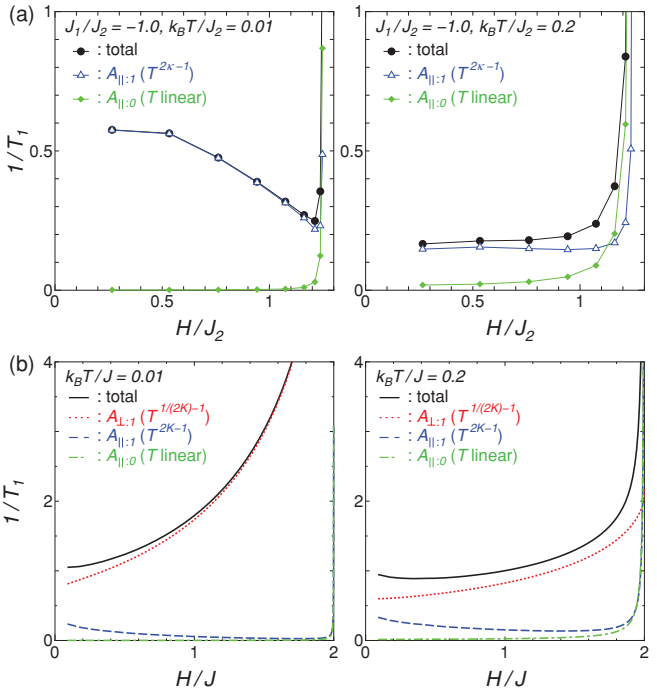

FIG. 4. (Color online) Field dependence of NMR relaxation rate $1/T_1$ for (a) the magnetic quadrupolar TL-liquid phase in the J_1-J_2 spin chain (1) with $J_1/J_2 = -1.0$ and (b) the standard TL-liquid phase in the spin-$\frac{1}{2}$ Heisenberg chain (12). Left (right) panels show the data for $k_BT/J_2 = 0.01$ and $k_BT/J = 0.01$ ($k_BT/J_2 = 0.2$ and $k_BT/J = 0.2$). In addition to the total value of $1/T_1$, each term in Eqs. (11) and (15) is shown separately.

Fig. 3. It indicates that $1/T_1$ is a monotonically increasing (decreasing) function of H (T) in the TL-liquid phase of the model (12) for sufficiently low temperatures. These two properties are completely different from those of the nematic TL-liquid phase shown in Figs. 1 and 2. The relaxation rate of real quasi-1D spin-$\frac{1}{2}$ AF magnets has been observed in the somewhat restricted regime of T and H, for example, in Refs. 46–49. Their results seem to agree nicely with our results in Fig. 3.

Finally, let us separately estimate each term in the expressions of $1/T_1$, Eqs. (11) and (15). They are depicted in Fig. 4. We find that in the Heisenberg chain (12), the relaxation rate $1/T_1$ is clearly governed by the contribution from the transverse spin correlation, that is, the $A_{\perp:1}$ term. In the nematic case, in which the transverse spin fluctuation is gapped, the $A_{\|:1}$ term becomes dominant in a wide magnetic-field range and is responsible for the behavior of $1/T_1$ decreasing with field H. This figure definitely demonstrates that characteristic features of $1/T_1$ in the J_1-J_2 spin chain originate from the absence of gapless modes in the transverse spin excitations.

From these results, we conclude that the NMR relaxation rates $1/T_1$ in the nematic and standard TL-liquid phases exhibit quite different behavior in T-H space, and hence we propose that its T and H dependence can be used to distinguish the nematic liquid phase from the standard TL liquid. Similar characteristic behavior of $1/T_1$ is also expected in the higher-order multipolar liquid phases in the J_1-J_2 spin chain, since these phases share essentially the same properties of spin correlations and the field H dependence of the TL-liquid parameter. Furthermore, a nematic TL-liquid phase in the SDW regime ($\kappa > 1/2$) is also shown to appear in a wide parameter region of the J_1-J_2 chain with AF J_1.[45,50] This AF-J_1 SDW$_2$ phase, where the transverse spin correlation is short-ranged, is also expected to show a peculiar H dependence of $1/T_1$, which is distinct from that in the usual TL liquid.

V. WIDE TEMPERATURE RANGE, HYPERFINE COUPLINGS, AND DM INTERACTIONS

So far we have discussed low-temperature behavior in spin isotropic pure 1D systems. In this section, we consider the temperature dependence of $1/T_1$ in a wide temperature range, taking interchain couplings into account. We also discuss the relation between the form of hyperfine coupling tensor $\mathcal{A}_{\mu\nu}$ and the direction of field H, as well as the effects of Dzyaloshinsky-Moriya (DM) interactions.

A. Temperature dependence of $1/T_1$ in a wide temperature range

First we consider a wide temperature window including higher and lower temperatures in which simple effective Hamiltonians (8) and (13) are no longer valid for describing

the physics of real quasi-1D magnets. At sufficiently low temperature, which is lower than the energy scale of weak 3D interchain couplings J_{3D}, long-range-ordered phases usually emerge due to 3D couplings and hence 1D effective theories cannot be applicable. In the case of the SDW regime, $1/T_1$ exhibits a divergence with a critical exponent near the critical temperature $\sim 1/(T-T_c)^{\xi}$, if 3D ordering occurs through a continuous (second-order) transition. In the case of the nematic regime, the nematic ordering at finite temperature may induce singularity in $1/T_1$. On the other hand, at high temperature, the effective TL-liquid theory becomes unreliable when the energy scale k_BT is increased up to the order of u/a, which is of the order of the 1D couplings J_1 and J_2, except for the saturation limit $H \to H_c$, where $u \to 0$. Deviation from the TL-liquid theory also comes from breaking of magnon bound states at high temperatures. The binding energy[51] is numerically estimated, at most, as $E_{\rm bind} \simeq 0.39J_2$, $0.28J_2$, and $0.14J_2$ for $J_1/J_2 = -2.0$, -1.0, and -0.6, respectively, which gradually decreases with lowering magnetization and vanishes at the border to the lower-field vector chiral phase. Thus, our prediction is valid in the temperature range $J_{3D} \ll k_BT \ll J_{1D} = \min(u/a, E_{\rm bind})$. In the higher-temperature regime $k_BT \gg J_{1D}$, the relaxation rate converges to a constant value.[52] We draw schematic patterns of the temperature dependence of $1/T_1$ in the nematic TL liquid in Figs. 5(a) and 5(b).

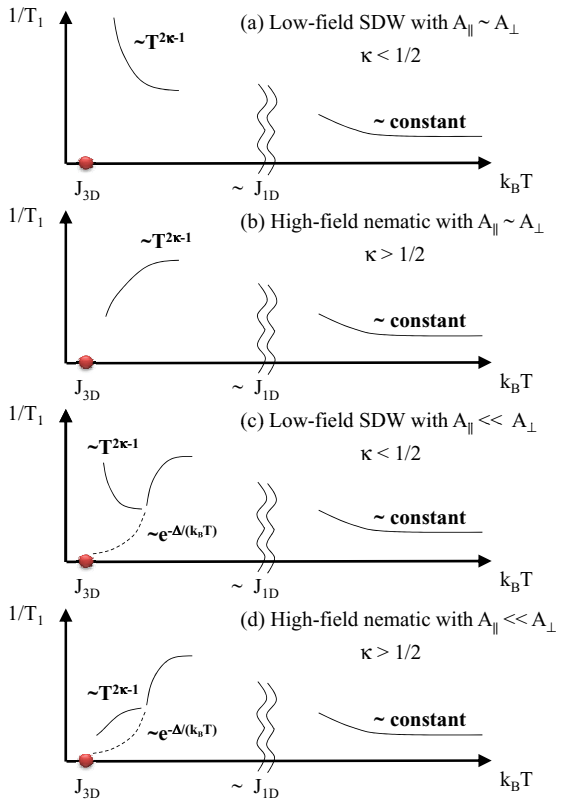

FIG. 5. (Color online) Schematic behavior of $1/T_1$ of the nematic TL liquid in a wide temperature region for several parameter settings. In the regime $J_{3D} \ll k_BT \ll J_{1D}$, our prediction based on low-energy effective theories can be applicable.

B. Hyperfine coupling tensor and field direction

Next we discuss the anisotropic case $A_\perp \neq A_\parallel$ (we have assumed $A_{\perp:n} = A_{\parallel:n}$ so far for simplicity). In the extreme case $A_\perp \gg A_\parallel$, we can no longer neglect the contribution from the exponentially decaying transverse spin correlation in $1/T_1$. In this extreme case, power-law behavior in $1/T_1$ of multipolar TL liquids is negligible down to extremely low temperature, hence we observe a thermal activation form $\sim e^{-\Delta/(k_BT)}$ in $1/T_1$ in a low-temperature regime $k_BT \ll J_{1D}$. Thus it is easy to detect a characteristic feature of multipolar TL liquids. If target magnets have sufficiently high crystallographic symmetry, the principal axes for hyperfine coupling tensor $\mathcal{A}_{\mu\nu}$ can be defined and the tensor is diagonalized. In this case, tuning the field direction parallel to a principal axis, we can eliminate off-diagonal elements $\mathcal{A}_{xz}$ and $\mathcal{A}_{yz}$, and equivalently set $A_\parallel = 0$. Therefore, the setup of H parallel to the axis offers an easy way of measuring the transverse spin gap and distinguishing multipolar liquids from ordinary TL liquid. Figures 5(c) and 5(d) present the schematic temperature dependence of $1/T_1$ in the nematic and SDW$_2$ TL liquids for the anisotropic case $A_\perp \gg A_\parallel$. In the opposite limit $A_\perp \ll A_\parallel$, our predictions in Figs. 1 and 2 are highly reliable in $J_{3D} \ll k_BT \ll J_{1D}$. However, we should note that in this limit $A_\perp \ll A_\parallel$, $1/T_1$ becomes insensitive to the disappearance of the algebraically decaying transverse spin correlation, the hallmark of the nematic liquid, and is not efficient in distinguishing the usual and nematic TL liquids.

C. Effects of the DM interaction

In the rest of this section, we consider the effects of magnetic anisotropies which generally exist in real magnets. In spin-$\frac{1}{2}$ systems, one of the most realistic anisotropies is the DM interaction defined by

$$\mathcal{H}_{\rm DM}(Q,\varphi,m) = \sum_j \cos(Qj+\varphi)\boldsymbol{D}\cdot(\boldsymbol{S}_j \times \boldsymbol{S}_{j+m}) \qquad (17)$$

for $m = 1,2$. Possible values of Q, φ, and the DM vector $\boldsymbol{D}$ depend strongly on the crystal structure of each compound. Since the DM interaction can sometimes induce an excitation gap accompanied with local spin polarization, it may violate the multipolar TL liquids. We discuss the effects of the DM interaction on the nematic TL liquid in the low-energy theory.

In the weak-coupling theory[8,10,53] for the spin-$\frac{1}{2}$ J_1-J_2 chain with $|J_1| \ll J_2$, the nematic phase is expressed by the effective Hamiltonian

$$\tilde{\mathcal{H}}^{\rm eff}_{J_1-J_2} = \int dy \sum_{\gamma=\pm} \frac{v_\gamma}{2}\left[K_\gamma^{-1}(\partial_y\phi_\gamma)^2 + K_\gamma(\partial_y\theta_\gamma)\right] + c_1(2a)^{-1}\sin(\sqrt{8\pi}\phi_- + \pi M) + c_2(\partial_y\theta_+)\sin(\sqrt{2\pi}\theta_-) + \cdots \qquad (18)$$

under the condition that only the c_1 term is relevant to the Gaussian model, where $y = 2ja$, $(\phi_\pm,\theta_\pm) = (\phi_1 \pm \phi_2, \theta_1 \pm \theta_2)/\sqrt{2}$, and $(\phi_{1(2)},\theta_{1(2)})$ is a boson-field pair defined in each of the decoupled AF-J_2 chains (see Sec. III B). The coupling constants $c_{1,2}$ are proportional to J_1. The c_1 term makes ϕ_- pinned. As a result, the transverse spin correlation decays in an exponential fashion. The remaining (ϕ_+,θ_+) sector

induces the gapless behavior of longitudinal-spin and nematic correlation functions. On the other hand, the c_2 term is known to induce a vector chiral phase[53–55] with $\langle(\boldsymbol{S}_j \times \boldsymbol{S}_{j+1})^z\rangle \sim \langle\sin(\sqrt{2\pi}\theta_-)\rangle \neq 0$ in a lower-field regime.[8,53] Let us concentrate on the former nematic TL liquid in the following.

Using the effective theory (18), we investigate five typical situations: $\mathcal{H}^{\rm i}_{\rm DM} = \mathcal{H}_{\rm DM}(0,0,1)$, $\mathcal{H}^{\rm ii}_{\rm DM} = \mathcal{H}_{\rm DM}(\pi,0,1)$, $\mathcal{H}^{\rm iii}_{\rm DM} = \mathcal{H}_{\rm DM}(0,0,2)$, $\mathcal{H}^{\rm iv}_{\rm DM} = \mathcal{H}_{\rm DM}(\pi,0,2)$, and $\mathcal{H}^{\rm v}_{\rm DM} = \mathcal{H}_{\rm DM}(\pi/2,\pi/4,2)$. Let us first consider the case of $\boldsymbol{D} = (0,0,D_z)$, that is, the DM vector is parallel to the applied field H. In this case, we find that the bosonized DM couplings $\mathcal{H}^{\rm i-iv}_{\rm DM}$ contain slowly moving bosonic terms without oscillating factors $(-1)^j$ or $e^{iq\pi Mj}$ (q is an integer). They are represented as

$$\mathcal{H}^{\rm i}_{\rm DM} \sim D_z \sin(\sqrt{2\pi}\theta_-) + \cdots, \tag{19a}$$

$$\mathcal{H}^{\rm ii}_{\rm DM} \sim D_z \Big\{ \cos(\sqrt{2\pi}\theta_-)[2a\sqrt{2\pi}\,\partial_y\theta_- + \cdots] - \sin(\sqrt{2\pi}\theta_-)\Big[(2a)^2\frac{\pi}{2}((\partial_y\theta_+)^2 + (\partial_y\theta_-)^2) + \cdots\Big]\Big\}, \tag{19b}$$

$$\mathcal{H}^{\rm iii}_{\rm DM} \sim D_z\sqrt{\pi}(2a)\partial_y\theta_+ + \cdots, \tag{19c}$$

$$\mathcal{H}^{\rm iv}_{\rm DM} \sim D_z\sqrt{\pi}(2a)\partial_y\theta_- + \cdots. \tag{19d}$$

In general, DM terms with $\boldsymbol{D} = (0,0,D_z)$ are invariant under a global U(1) spin rotation $S^+_j \to S^+_j e^{i\varphi}$, which corresponds to a shift of the phase field $\theta_+ \to \theta_+ + \sqrt{2/\pi}\varphi$. Therefore, the bosonized DM terms with D_z do not involve any vertex operator with θ_+. In addition, all the translationally symmetric DM terms do not contain any vertex term with ϕ_+ at least for the case of incommensurate values of M, since the $2n$-site translation induces $\phi_{1,2}(y) \to \phi_{1,2}(y+2na) + n\sqrt{\pi}(1/2+M)$ and equivalently $\phi_+(y) \to \phi_+(y+2na) + n\sqrt{\pi/2}(1+2M)$. Equation (19) is consistent with these symmetry arguments. Two DM interactions $\mathcal{H}^{\rm i,ii}_{\rm DM}$ tend to lock θ_- together with the c_2 term in Eq. (18), both of which compete with the c_1 term. In the nematic phase, we know that the c_1 term is most relevant and hence the DM interactions $\mathcal{H}^{\rm i,ii}_{\rm DM}$ with sufficiently small D_z are negligible. In the strong DM coupling regime, these DM terms compete with the c_1 term and could change the nematic liquid into a chiral phase with $\langle(\boldsymbol{S}_j \times \boldsymbol{S}_{j+1})^z\rangle \neq 0$. On the other hand, the leading terms of $\mathcal{H}^{\rm iii,iv}_{\rm DM}$ can be absorbed into the free-boson part by shifting the field $\theta_\pm$. The shift brings about a finite expectation value $\langle(\boldsymbol{S}_j \times \boldsymbol{S}_{j+2})^z\rangle \sim \langle\partial_y\theta_{1,2}\rangle$, but we note that these chiralities do not accompany any spontaneous symmetry breaking. The shift also affects the form of spin dynamical structure factors, that is, the gapless points of $\mathcal{S}^{\pm\mp}(k,\omega)$ are slightly changed and a small asymmetry of k dependence emerges. In contrast to $\mathcal{H}^{\rm i,ii}_{\rm DM}$, $\mathcal{H}^{\rm iii,iv}_{\rm DM}$ do not compete with the c_1 term. Therefore, chirality and nematic quasi-long-range order can coexist in the latter case of $\mathcal{H}^{\rm iii,iv}_{\rm DM}$. This symmetry argument indicates that the DM terms with D_z do not introduce any vertex operator in the (ϕ_+,θ_+) sector, and this statement would be true in the wide nematic-liquid region regardless of the value of J_1. The gapless nature of the (ϕ_+,θ_+) sector hence survives even after introducing these DM terms. From these discussions, we conclude that the nematic TL liquid survives even in the presence of DM terms with $\boldsymbol{D} = (0,0,D_z)$ at least when $D_z \ll |J_{1,2}|$. Thus, the nematic liquid is relatively stable against DM interactions with $\boldsymbol{D} = (0,0,D_z)$.

For the case of $\boldsymbol{D} = (D_x,0,0)$, the U(1) rotational symmetry is broken and the emergence of vertex operators $e^{iq\sqrt{\pi}\theta_+}$ is generally allowed. In fact, $\mathcal{H}^{\rm v}_{\rm DM}$ is bosonized as

$$\mathcal{H}^{\rm v}_{\rm DM} \sim D_x M \sin(\sqrt{\pi/2}\theta_+)\cos(\sqrt{\pi/2}\theta_-) + \cdots. \tag{20}$$

This leading term has scaling dimension $1/(8K_+) + 1/(8K_-)$ and can generate a staggered magnetization along the S^y axis in each AF-J_2 chain. In the weak J_1 limit, $\mathcal{H}^{\rm v}_{\rm DM}$ is more relevant than the c_1 term with scaling dimension $2K_- \approx 2K$ and it can violate the nematic liquid phase. [In this limit, $K_\pm$ approaches the value of original TL-liquid parameter K ($1/2 < K < 1$) of the AF-J_2 chain in magnetic field.] When J_1 is sufficiently strong, the c_1 term defeats the perturbation $\mathcal{H}^{\rm v}_{\rm DM}$ and protects the nematic liquid phase, pinning ϕ_-. In this case, $\sin(\sqrt{\pi/2}\theta_+)\cos(\sqrt{\pi/2}\theta_-)$ generates new operators $\cos(\sqrt{2\pi}\theta_+)$ and $\cos(\sqrt{2\pi}\theta_-)$ via the renormalization-group process. The first term with scaling dimension $1/(2K_+)$ can open a gap in the (ϕ_+,θ_+) sector and induce a transverse staggered magnetization, since the U(1) spin symmetry is broken by the DM term. In the weak DM coupling regime, thus, this DM term perturbatively deforms nematic spin liquid, opening a small gap in longitudinal spin modes and inducing a small expectation value of transverse spins. The other DM interactions $\mathcal{H}^{\rm i-iv}_{\rm DM}$ with $\boldsymbol{D}$ parallel to the x axis do not possess any slowly moving operator within a naive calculation based on the bosonization, but they generally have the ability to generate $e^{iq\sqrt{\pi}\theta_+}$, which is allowed from the symmetry argument. Thus, the gapless nature of the (ϕ_+,θ_+) sector is expected to be fragile and unstable against DM terms with $\boldsymbol{D} \neq (0,0,D_z)$.

These discussions on DM terms indicate that if we apply magnetic field parallel to the DM vector $\boldsymbol{D}$, the nematic TL liquid is stably realized and our prediction of $1/T_1$ is reliable in wider H-T space. Even when $\boldsymbol{D}$ is not parallel to H, the nematic phase will also survive if the DM coupling is small, $|J_{1,2}| \gg |\boldsymbol{D}|$, and if the system is in the temperature regime $k_BT \gg |\boldsymbol{D}|$.

VI. CONCLUSIONS

We have evaluated accurately the field and temperature dependence of the NMR relaxation rate $1/T_1$ in magnetic quadrupolar (spin nematic) TL liquid of the spin-$\frac{1}{2}$ J_1-J_2 chain, combining field-theoretical techniques with DMRG results (see Figs. 1 and 2). As a comparison, we have also calculated $1/T_1$ of the spin-$\frac{1}{2}$ AF Heisenberg chain, using field theories, the Bethe ansatz, and the DMRG method (see Fig. 3).

In the nematic and SDW_2 TL-liquid phase at a low temperature $k_BT \ll |J_{1,2}|$, the relaxation rate $1/T_1$ first decreases with increasing magnetic field and then rapidly increases near saturation. In the higher-temperature regime $k_BT \sim 0.1J_2$, the field dependence of $1/T_1$ becomes quite small except in the vicinity of saturation. This nonmonotonic behavior clearly differs from the monotonically increasing behavior in the usual TL liquid in spin-$\frac{1}{2}$ AF chains. The decreasing behavior of $1/T_1$ with increasing H comes from the monotonic

increase in the TL-liquid parameter κ, while the rapid increase in $1/T_1$ near saturation is attributed to the decrease of velocity $u \to 0$. The monotonic property of κ is also essential for the characteristic temperature dependence[15] of $1/T_1$: with lowering temperature, $1/T_1$ increases in an algebraic form in the lower-field SDW regime ($\kappa < 1/2$), whereas it decreases in the higher-field nematic regime ($\kappa > 1/2$). These characteristic H and T dependences could be a signature of the nematic and SDW_2 TL-liquid phase. Similar features are also expected to appear in higher-order multipolar TL liquids, for example, octupolar and hexadecapolar TL liquids. Probing the H dependence would be easier than doing the T dependence since the former does not necessarily require the accession to the high-field regime, where NMR measurements are difficult. A combination of the H and T dependences of $1/T_1$ and the gapless behavior observed from bulk quantities (such as specific heat and magnetic susceptibilities) would present indirect but strong evidence for multipolar TL-liquid phases.

In Sec. V, we also considered the effects of spin anisotropies and interchain couplings, which are neglected in the ideal J_1-J_2 spin-chain model. In particular, we point out that if the direction of the external field H can be parallel to the principal axis of the hyperfine-coupling tensor, all the algebraic contributions in the temperature dependence of $1/T_1$, that is, $A_{\parallel:n}$ terms, disappear. As a result, $1/T_1$ of multipolar phases in the J_1-J_2 chain becomes a thermal activation form $\sim e^{-\Delta/(k_B T)}$. In addition, we predict that the nematic TL liquid is stable for small DM terms with the DM vector parallel to the applied field, while it can be easily deformed by the DM terms, thereby accompanied with small transverse staggered magnetization, when the DM vector is perpendicular to the field. This indicates that we should apply magnetic field parallel to the DM vector to obtain a stable nematic liquid phase in real quasi-1D J_1-J_2 magnets.

Finally, we comment on some real compounds. Recently, quasi-1D edge-sharing cuprate magnets, for example, $LiCu_2O_2$, $LiCuVO_4$, $Rb_2Cu_2Mo_3O_{12}$, and $PbCuSO_4(OH)_2$, have been studied extensively as low-dimensional frustrated or multiferroic systems. Their magnetic properties are believed to be described by spin-$\frac{1}{2}$ J_1-J_2 chains in a certain temperature regime. Except for $Rb_2Cu_2Mo_3O_{12}$, a 3D ordering has been observed below a very low critical temperature T_c at least in zero magnetic field. Furthermore, the values of J_1 and J_2 have been semiquantitatively estimated in several ways. The estimated coupling constants and the critical temperatures in $LiCu_2O_2$ (Refs. 25–28), $PbCuSO_4(OH)_2$ (Refs. 21–24), and $Rb_2Cu_2Mo_3O_{12}$ (Ref. 20) are, respectively, $(J_1/k_B, J_2/k_B, T_c) \sim (-138\ \mathrm{K}, 86\ \mathrm{K}, 24\ \mathrm{K})$, $\sim (-13\ \mathrm{K}, 21\ \mathrm{K}, 2.8\ \mathrm{K})$, and $(J_1/k_B, J_2/k_B) \sim (-138\ \mathrm{K}, 51\ \mathrm{K})$, while two different results have been reported for $LiCuVO_4$ (Refs. 16–19 and 56): $(J_1/k_B, J_2/k_B, T_c) \sim (-19\ \mathrm{K}, 45\ \mathrm{K}, 2\ \mathrm{K})$ and $\sim(-182\ \mathrm{K}, 91\ \mathrm{K}, 2\ \mathrm{K})$. The critical temperatures T_c are thus small compared to the magnitude of $J_{1,2}$ except for $LiCu_2O_2$. This means that three dimensionality is small in these compounds. We note that our prediction for $1/T_1$ could be applied to the temperature condition $k_B T_c \ll k_B T \ll |J_{1,2}|$.

Among these compounds, the magnetization process of $LiCuVO_4$ has been intensively studied in some experimental groups. In the low-field regime including zero field, spiral phases exist at low temperatures. Above $H \approx 7.5$ T, the spiral phase turns into another phase, which was concluded by NMR measurements[19] to be a modulated collinear phase. The appearance of both the spiral and modulated collinear phases was well understood[8] as a consequence of the vector chiral phase and the incommensurate SDW_2 phase in the 1D J_1-J_2 spin chain. In addition, quite recently, a new phase transition has been observed[17] at $H \approx 40$ T, where the saturation field is $H_s \approx 47$ T. Comparing the result with the phase diagram in the 1D J_1-J_2 spin chain,[8,11] we expect the new high-field phase for $40 < H < 47$ T to be a nematic long-range-ordered phase. So far, the magnetic structure of this new phase has not been experimentally identified at all. We expect that NMR measurements above the critical temperatures of this new high-field phase and of the intermediate-field modulated collinear phase would be useful to verify whether the new phase is a nematic phase. We also note that $PbCuSO_4(OH)_2$ has a rather small saturation field $H_s \approx 10$ T.[24] This might be an ideal material for measuring the H dependence of $1/T_1$ up to the saturation field.

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[1] A. F. Andreev and I. A. Grishchuk, Sov. Phys. JETP **60**, 267 (1984).
[2] X. G. Wen, F. Wilczek, and A. Zee, Phys. Rev. B **39**, 11413 (1989).
[3] A. V. Chubukov, Phys. Rev. B **44**, 4693 (1991).
[4] T. Momoi and N. Shannon, Prog. Theor. Phys. Suppl. **159**, 72 (2005).
[5] N. Shannon, T. Momoi, and P. Sindzingre, Phys. Rev. Lett. **96**, 027213 (2006).
[6] T. Momoi, P. Sindzingre, and N. Shannon, Phys. Rev. Lett. **97**, 257204 (2006).
[7] L. Kecke, T. Momoi, and A. Furusaki, Phys. Rev. B **76**, 060407(R) (2007).
[8] T. Hikihara, L. Kecke, T. Momoi, and A. Furusaki, Phys. Rev. B **78**, 144404 (2008).
[9] R. Shindou and T. Momoi, Phys. Rev. B **80**, 064410 (2009).
[10] T. Vekua, A. Honecker, H.-J. Mikeska, and F. Heidrich-Meisner, Phys. Rev. B **76**, 174420 (2007).
[11] J. Sudan, A. Luscher, and A. M. Läuchli, Phys. Rev. B **80**, 140402(R) (2009).
[12] H. T. Ueda and K. Totsuka, Phys. Rev. B **80**, 014417 (2009).
[13] S. Nishimoto, S.-L. Drechsler, R. O. Kuzian, J. van den Brink, J. Richter, W. E. A. Lorenz, Y. Skourski, R. Klingeler, and B. Buechner, e-print arXiv:1004.3300.
[14] M. E. Zhitomirsky and H. Tsunetsugu, Europhys. Lett. **92**, 37001 (2010).
[15] M. Sato, T. Momoi, and A. Furusaki, Phys. Rev. B **79**, 060406(R) (2009).

[16]M. Enderle, C. Mukherjee, B. Fåk, R. K. Kremer, J.-M. Broto, H. Rosner, S.-L. Drechsler, J. Richter, J. Malek, A. Prokofiev, W. Assmus, S. Pujol, J.-L. Raggazzoni, H. Rakoto, M. Rheinstädter, and H. M. Rønnow, Europhys. Lett. **70**, 237 (2005).

[17]L. E. Svistov, T. Fujita, H. Yamaguchi, S. Kimura, K. Omura, A. Prokofiev, A. I. Smirnov, Z. Honda, and M. Hagiwara, JETP Lett. **93**, 24 (2011); e-print arXiv:1005.5668 .

[18]Y. Naito, K. Sato, Y. Yasui, Y. Kobayashi, Y. Kobayashi, and M. Sato, J. Phys. Soc. Jpn. **76**, 023708 (2007).

[19]N. Büttgen, H.-A. Krug von Nidda, L. E. Svistov, L. A. Prozorova, A. Prokofiev, and W. Assmus, Phys. Rev. B **76**, 014440 (2007).

[20]M. Hase, H. Kuroe, K. Ozawa, O. Suzuki, H. Kitazawa, G. Kido, and T. Sekine, Phys. Rev. B **70**, 104426 (2004).

[21]G. Kamieniarz, M. Bieliński, G. Szukowski, R. Szymczak, S. Dyeyev, and J. -P. Renard, Comput. Phys. Commun. **147**, 716 (2002).

[22]M. Baran, A. Jedrzejczak, H. Szymczak, V. Maltsev, G. Kamieniarz, G. Szukowski, C. Loison, A. Ormeci, S.-L. Drechsler, and H. Rosner, Phys. Status Solidi (c) **3**, 220 (2006).

[23]Y. Yasui *et al.*, submitted to J. Phys. Soc. Jpn. (unpublished).

[24]A. U. B. Wolter, R. Vogel, Y. Skourski, K. C. Rule, S. Süllow, G. Heide, and B. Buechner, International Conference on Magnetic Resonance in Highly Frustrated Magnetic Systems, Slovenia, (2010).

[25]T. Masuda, A. Zheludev, A. Bush, M. Markina, and A. Vasiliev, Phys. Rev. Lett. **92**, 177201 (2004).

[26]T. Masuda, A. Zheludev, B. Roessli, A. Bush, M. Markina, and A. Vasiliev, Phys. Rev. B **72**, 014405 (2005).

[27]S. Park, Y. J. Choi, C. L. Zhang, and S-W. Cheong, Phys. Rev. Lett. **98**, 057601 (2007).

[28]S. Seki, Y. Yamasaki, M. Soda, M. Matsuura, K. Hirota, and Y. Tokura, Phys. Rev. Lett. **100**, 127201 (2008).

[29]S.-L. Drechsler, J. Richter, A. A. Gippius, A. Vasiliev, A. A. Bush, A. S. Moskvin, J. Málek, Yu. Prots, W. Schnelle, and H. Rosner, Europhys. Lett. **73**, 83 (2006).

[30]C. P. Slichter, *Principles of Magnetic Resonance* (Springer-Verlag, Berlin, 1990).

[31]T. Goto, T. Ishikawa, Y. Shimaoka, and Y. Fujii, Phys. Rev. B **73**, 214406 (2006).

[32]In the narrow range $-2.720 < J_1/J_2 < -2.669$, the soft two-magnon bound state has an incommensurate momentum $k \neq \pi$ to realize a two-component TL liquid. In this paper, we do not consider this case and focus on the one-component multipolar TL liquids of the multimagnon bound states with momentum $k = \pi$. However, it is naively expected that the incommesurate multipolar liquid qualitatively shares the features of $1/T_1$ in the commensurate multipolar liquids (see Secs. III and IV) which mainly result from the formation of multimagnon bound states.

[33]F. D. M. Haldane, Phys. Rev. Lett. **47**, 1840 (1981).

[34]T. Giamarchi, *Quantum Physics in One Dimension* (Oxford University Press, New York, 2004).

[35]R. Chitra and T. Giamarchi, Phys. Rev. B **55**, 5816 (1997).

[36]T. Giamarchi and A. M. Tsvelik, Phys. Rev. B **59**, 11398 (1999).

[37]M. Klanjšek, H. Mayaffre, C. Berthier, M. Horvatić, B. Chiari, O. Piovesana, P. Bouillot, C. Kollath, E. Orignac, R. Citro, and T. Giamarchi, Phys. Rev. Lett. **101**, 137207 (2008).

[38]P. Bouillot, C. Kollath, A. M. Läuchli, M. Zvonarev, B. Thielemann, C. Rüegg, E. Orignac, R. Citro, M. Klanjšek, C. Berthier, M. Horvatić, and T. Giamarchi, e-print arXiv:1009.0840.

[39]N. M. Bogoliubov, A. G. Izergin, and V. E. Korepin, Nucl. Phys. B **275**, 687 (1986).

[40]S. Qin, M. Fabrizio, L. Yu, M. Oshikawa, and I. Affleck, Phys. Rev. B **56**, 9766 (1997).

[41]D. C. Cabra, A. Honecker, and P. Pujol, Phys. Rev. B **58**, 6241 (1998).

[42]For instance, the list of the values of K and v are summarized in F. H. L. Essler, A. Furusaki, and T. Hikihara, Phys. Rev. B **68**, 064410 (2003).

[43]T. Hikihara and A. Furusaki, Phys. Rev. B **69**, 064427 (2004); the detailed list of a_1 and b_0 is presented in e-print arXiv:0310391[cond-mat].

[44]There are logarithmic corrections to Eq. (15) near the zero-field SU(2)-symmetric point with $K = 1/2$, which we omitted. See S. Sachdev, Phys. Rev. B **50**, 13006 (1994); M. Takigawa, N. Motoyama, H. Eisaki, and S. Uchida, Phys. Rev. Lett. **76**, 4612 (1996); M. Takigawa, O. A. Starykh, A. W. Sandvik, and R. R. P. Singh, Phys. Rev. B **56**, 13681 (1997).

[45]T. Hikihara, T. Momoi, A. Furusaki, and H. Kawamura, Phys. Rev. B **81**, 224433 (2010).

[46]L. J. Azevedo, A. Narath, Peter M. Richards, and Z. G. Soos, Phys. Rev. Lett. **43**, 875 (1979).

[47]L. J. Azevedo, A. Narath, P. M. Richards, and Z. G. Soos, Phys. Rev. B **21**, 2871 (1980).

[48]J. P. Groen, T. O. Klaassen, N. J. Poulis, G. Müller, H. Thomas, and H. Beck, Phys. Rev. B **22**, 5369 (1980).

[49]H. Kühne, M. Günther, S. Grossjohann, W. Brenig, F. J. Litterst, A. P. Reyes, P. L. Kuhns, M. M. Turnbull, C. P. Landee, and H.-H. Klauss, Phys. Status Solidi B **247**, 671 (2010).

[50]K. Okunishi and T. Tonegawa, J. Phys. Soc. Jpn. **72**, 479 (2003). In this paper, the nematic TL liquid is called the even-odd phase.

[51]The magnon binding energy is defined as follows. First we prepare $\Delta_n(\mathcal{M}) = E_0(\mathcal{M}+n) - E_0(\mathcal{M})$, where $E_0(\mathcal{M})$ is the lowest energy eigenvalue in the subspace with fixed $\sum_j S_j^z = \mathcal{M} = ML$ (L is the total number of sites). Calculating the binding energies of the two definitions $E_+ = 2\Delta_1 - \Delta_2$ and $E_- = 2\Delta_{-1} - \Delta_{-2}$, we obtain the true binding energy from the minimum value of them, $E_{\text{bind}} = \min(E_+, E_-)$.

[52]T. Moriya, Prog. Theor. Phys. **16**, 23 (1956); **16**, 641 (1956).

[53]A. Kolezhuk and T. Vekua, Phys. Rev. B **72**, 094424 (2005).

[54]A. A. Nersesyan, A. O. Gogolin, and F. H. L. Eßler, Phys. Rev. Lett. **81**, 910 (1998).

[55]T. Hikihara, M. Kaburagi, and H. Kawamura, Phys. Rev. B **63**, 174430 (2001).

[56]J. Sirker, Phys. Rev. B **81**, 014419 (2010).