Electron Spin Resonance in Quasi-One-Dimensional Quantum Antiferromagnets: Relevance of Weak Interchain Interactions

Shunsuke C. Furuya and Masahiro Sato

1 DPMC-MaNEP, University of Geneva, 24 Quai Ernest-Ansermet CH-1211 Geneva, Switzerland
2 Department of Physics and Mathematics, Aoyama Gakuin University, Sagamihara, Kanagawa 229-8558, Japan

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We argue universal features on the electron spin resonance (ESR) of Tomonaga-Luttinger liquid (TLL) phases in a wide class of weakly coupled $S = 1/2$ antiferromagnetic spin chains such as spin ladders, spin tubes and three-dimensionally coupled spin chains. We show that the ESR linewidth of coupled chains increases with lowering temperature while the linewidth of a single spin chain is typically proportional to temperature. This broadening is attributed to interchain interactions. We demonstrate that our theory can account for anomalous behaviors of the linewidths in an $S = 1/2$ four-leg spin tube compound Cu$_2$Cl$_4$:H$_8$C$_4$SO$_2$ (abbreviated to Sul-Cu$_2$Cl$_4$) and a three-dimensionally coupled $S = 1/2$ spin chain compound CuCl$_2$·2NC$_5$H$_5$.

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Introduction. — In condensed matter physics, various experimental techniques have been continuously developed. In particular, ways of observing responses to electromagnetic waves are utilized for understanding dynamical properties (e.g., elementary excitations) of target materials [1]. Among them, the electron spin resonance (ESR) is characteristic thanks to its high resolution and sensitivity to symmetries of interactions between electron spins [2, 3]. For instance, the SU(2) rotational symmetry manifests as a zero linewidth in ESR spectra (i.e., the ESR spectrum is delta-function-like) [4–6]. However, in actual compounds, the SU(2) symmetry is more or less broken due to, for instance, dipolar interactions. Such anisotropic interactions generally induce a finite linewidth. ESR enables us to detect small anisotropic interactions which are hard to be observed by other experimental techniques [7].

The above argument indicates that a careful treatment of the SU(2) symmetry is essential to develop ESR theories. Classical theories for ESR in ordered magnets, in which the SU(2) symmetry is spontaneously broken, have been established [8]. On the other hand, in quantum liquid phases such as Tomonaga-Luttinger liquids (TLLs) [9] in quasi one-dimensional (1D) magnets and quantum spin liquids [10] in frustrated magnets, ESR is less understood. This is because the SU(2) symmetry is only weakly broken by small anisotropic interactions and even the dominant (i.e., SU(2)-symmetric) part is strongly correlated [10, 11]. On the experimental side, many intriguing ESR experiments in quantum disordered phases of quasi-1D magnets have been reported [12–13]. Moreover, in view of recent intensive experimental studies on spin ladders (Fig. 1 (a)) [14, 15] and spin tubes (Fig. 1 (b,c)) [20, 23], quantum theories for ESR are strongly called for.

A prominent development of the ESR theory for strongly interacting spin systems has been given by Oshikawa and Affleck [6]. They have developed a general theory for ESR spectra of single-component TLLs in $S = 1/2$ Heisenberg antiferromagnetic (AF) chains subject to anisotropic exchange and staggered Dzyaloshinskii-Moriya interactions along the chain. Note that TLL can deal with strong spin-spin interactions nonperturbatively [6]. Oshikawa-Affleck theory [6] successfully explained several ESR spectra of spin chain compounds. However, in addition to the chain direction, magnetic anisotropies usually exist along the interchain directions as well (Fig. 1). Unfortunately, at the moment, only a few studies considered effects of interchain interactions on ESR [2, 24]. In this paper, we therefore study general effects of interchain interactions on the ESR linewidth in quasi-1D quantum antiferromagnets. Our theory predicts a universal broadening of the ESR spectrum with lowering temperature $T$ in a wide class of quasi-1D antiferromagnets in Fig. 1. We point out that the broadening can (cannot) be explained by taking anisotropic interchain (intrachain) interactions into account. Temperature dependences of the linewidth are summarized in Fig. 2 and Table I. In a latter part of this paper, we will apply our theory to ESR experimental results of a four-leg spin tube compound Cu$_2$Cl$_4$:H$_8$C$_4$SO$_2$ (abbreviated to Sul-Cu$_2$Cl$_4$) [12, 22, 28] and a quasi-1D anti-
ferromagnet CuCl$_2$·2NC$_5$H$_5$ 29. These two materials are believed to possess a weak interchain interaction. We demonstrate that our theory describe the experiments on these magnets well.

**Two-Leg Spin Ladders.** — Let us first consider an $S = 1/2$ two-leg AF spin ladder in order to explicitly show the broadening by the interchain interaction. It is easy to extend the theory for the ladder to other quasi-1D magnets. The spin-ladder Hamiltonian is given by

$$H_{\text{ladder}} = \sum_{n=1,2} H_n + H_{\text{rung}} + H'. \quad (1)$$

The first term $H_n = J \sum_j S_{j,n} \cdot S_{j+1,n} - g \mu_B H \sum_j S_{j,n}^z$ is the $n$th-leg Heisenberg Hamiltonian, where $S_{j,n}$ is the spin operator at a site $j$ on a $n$th leg, $J > 0$ is the AF coupling constant, $g$ is the Landé factor, $\mu_B$ is the Bohr magneton, and $H$ is the external magnetic field. The term $H_{\text{rung}}$ denotes the exchange interaction along the interchain (rung) direction, $H_{\text{rung}} = J_r \sum_j S_{j,1} \cdot S_{j,2}$, in which $|J_r|$ is assumed to be small compared to $J$.

The term $H'$ is a weak anisotropic interaction to violate the SU(2) symmetry which induces a finite width. We employ a unit $\hbar = k_B = g \mu_B = 1$ in the following. From the field-theory approach 30-32, the low-energy physics of each leg is described by a TLL $H_n = \frac{\pi}{2} \int dx \left( (\partial_x \theta_n)^2 + (\partial_x \phi_n)^2 \right)$ where $x$ is the continuous coordinate, $(\phi_n, \theta_n)$ is the canonical pair of boson fields and $v \sim \pi J/2$ is the “Fermi” velocity. The spin operator is bosonized as $S_{j,n} = J_{nL} + J_{nR} + (-1)^j N_n. \quad (2)$

Here $J_n = J_{nL} + J_{nR}$ and $N_n$ are respectively the uniform and staggered components of the spin operator and $J_{nL(nR)}$ denotes its left-moving (right-moving) part. Explicit representations of bosonization formulas are not important, but they are given in the Supplemental Material 32 for self-containment.

When $H = 0$ and $H' = 0$, the relevant rung coupling,

$$H_{\text{rung}} = J_r \int dx (J_{1L} \cdot J_{2R} + J_{1R} \cdot J_{2L} + N_1 \cdot N_2) \quad (3)$$

generates a finite mass gaps $\Delta_1$ and $\Delta_\perp(\geq \Delta_1)$ in the triplet and singlet spin sectors, respectively 31. A weak anisotropy $H'$ slightly deforms the degeneracy of leg excitations. When $T > T_L \sim \Delta_1$, the rung coupling is negligible and the two-leg ladder can be regarded as two decoupled TLLs, while the TLL picture cannot be used in high temperature regime $T > T_H \sim J$. We will mainly consider the decoupled TLL phase in the range $T_L < T < T_H$ in a weak field region $H \ll T$.

According to the linear response theory, the ESR spectrum $I(\omega)$ is proportional to $-\omega \tilde{\text{Im}} G_{S_sS_s}(\omega, q = 0)$, in which $G_{S_sS_s}(\omega, q)$ is the retarded Green’s function of the transverse spins $S_{j=\alpha=\pm}(y)$, $\omega$ is the frequency of the external electromagnetic wave, and $q$ is the wave number. Here we assume that $H$ and the polarization of the electromagnetic wave are respectively set parallel to $S_z$ and $S^z$ or $y$ axes. The ESR spectrum probes the motion with $q = 0$. As far as we are in the TLL phase, we can easily show that the ESR spectrum of two decoupled TLLs has a Lorentzian line shape $\frac{1}{2}$ (see Fig. 2 (a)) near $\omega = H$, where the linewidth $\eta$ is given by $\frac{\beta}{2}$.

$$\eta = -\frac{1}{2 \langle \beta \rangle} \text{Im} G_{AA'}^{R}(\omega = H) \quad (4)$$

in the perturbative expansion with respect to the coupling constant of $H'$. Here $S^a = \sum_{j,\alpha} S_{\alpha,j}^a$ denotes a total spin and $A = [H', S^+]$. We note that $A$ is proportional to a small parameter of $H'$. In the retarded Green’s function $G_{AA'}^{R}(\omega) = -i \int_0^\infty dt e^{i\omega t} \langle [A(t), A(0)] \rangle$, the symbol $\langle \cdots \rangle_0$ denotes the average with respect to the unperturbed part $\text{Im} G_{AA'}(\omega = H) = \text{Im} G_{AA'}^{R}(\omega = H)$. When $H' = 0$, the linewidth is zero because $A$ trivially vanishes and the spin-spin interaction is isotropic. Let us consider a longitudinal rung anisotropy $H' = H'_L = J_r \delta_{Lz} \sum_j S_{j,1}^z S_{j,2}^z$ with $|\delta_{Lz}| \ll 1$. This anisotropic interaction generates

$$A = 2 J_r \delta_{Lz} (J_{Lz} + N_{Lz}), \quad (5)$$

where $J_{Lz}$ and $N_{Lz}$ are respectively the contributions from $J_n$ and $N_n$ in Eq. (5), and are expressed as

$$J_{Lz} = \int dx \sum_{n \neq \prime} \left( J_{nL}^2 : J_{nR}^2 R + J_{nR}^2 : J_{nL}^2 \right), \quad (6)$$

$$N_{Lz} = \int dx \left( N_{1z}^2 N_{2z}^z + N_{2z}^z N_{1z}^z \right). \quad (7)$$

Here $O := O - \langle O \rangle$ denotes a normal ordering. Specific representations of $J_{Lz}$ and $N_{Lz}$ are not important. The point is that $J_{Lz}$ and $N_{Lz}$ have conformal weights $(1, 1)$ and $(\frac{3}{2}, \frac{3}{2})$, respectively, in the decoupled TLL phase. The scaling dimension $d$ of an operator with a weight $(\Delta, \bar{\Delta})$ is given by $d = \Delta + \bar{\Delta}$. Hence, $J_{Lz}$ is marginal ($d = 2$) and $N_{Lz}$ is relevant ($d = 1$).

From the bosonization 31, a retarded Green’s function $G_{AA'}^{d(2,2)/d(2)}(\omega, q)$ of an operator with a weight $(d/2, d/2)$ at a finite $T$ can be computed as,

$$G_{\frac{d}{2}, \frac{d}{2}}^{R}(\omega, q) = -\sin(\pi d) F_d \left( \frac{\omega - vq}{4\pi T} \right) F_d \left( \frac{\omega + vq}{4\pi T} \right), \quad (8)$$

where $F_d(x) = (2\pi T/\nu)^{-1} B(\frac{x}{2} - i\nu, 1 - x)$ and $B(x, y)$ is the Beta function. The quantity $G_{AA'}^{R}(\omega = H)$ in Eq. (4) is written in terms of $G_{\Delta, \bar{\Delta}}^{R}(\omega, q)$ as

$$G_{AA'}^{R}(H) = 4J_r^2 \delta_{Lz} \left[ C_u C_s \left( \frac{\pi}{4} \right) \left( H - H^\perp \right) + C_s \left( \frac{\pi}{4} \right) \left( H - H^\perp \right) \right], \quad (9)$$

where $C_{u,s}$ are dimensionless constants that appear in the bosonization formula 31, 32. The representation 32 clearly shows the difference of our spin ladder and single spin chains with magnetic anisotropy.
can easily derive
\[ \text{Im} s \text{eg ladder is in the gapped phase with } \Delta \text{ as well, while the last term emerges due to } \] 

\[ \chi \text{ where } \] 

\[ \text{range } T > T_{	ext{cr}} (T < T_{L}) \text{, the linewidth is constant (gradually disappears down to } T \text{ )}. \] 

\[ \text{similar behavior of } \eta \text{ is also predicted to be observed in other quasi-1D AF magnets (see the text and Table I).} \]

in Ref. \[ \text{The first term of Eq. (9) comes from } J_{l} \text{ which is considered in the Oshikawa-Affleck theory} \] 

\[ \text{as well, while the last term emerges due to } N_{L} \text{ and it does not appear in the case of spin chains. One can easily derive } -\text{Im} G^{R}_{(1,1)}(H, H/v) = \pi^{2}HT/v^{2} \text{ and } -\text{Im} G^{R}_{(2,0)}(H, H/v) = \pi^{2}H/4T \text{ for } H/T < 1. \] 

\[ \eta = \frac{J_{l}^{2}S_{L}^{2}}{J_{X}} \frac{4C_{l}^{2}T}{\pi} + \frac{\pi^{2}C_{l}^{4}}{16} \frac{J_{l}^{2}}{T}, \] 

\[ \text{where } \chi = \langle S_{L}^{2} \rangle / H \text{ is the uniform susceptibility, which is insensitive to } T \text{ in the decoupled TLL phase. In the range } T_{L} < T < T_{H} \text{, there would exist the crossover temperature } T_{cr}. \] 

\[ \text{For } T > T_{cr} \text{, the first term } \propto T/J \text{ in Eq. (10) dominates the linewidth, while the second } \propto (T/J)^{-1} \text{ does it for } T < T_{cr}. \] 

\[ \text{Namely, the broadening } \eta \propto T^{-1} \text{ (the exchange narrowing } \eta \propto T) \text{ occurs when } T < T_{cr} (T > T_{cr}). \] 

\[ \text{It is worth noting that } T_{H} \text{ and } T_{cr} \text{ are both an order of } J, \text{ and thus the exchange narrowing region } T_{cr} < T < T_{H} \text{ is narrow or it does not exist (Fig. 2(b)).} \]

We argue the linewidth in regions where the decoupled TLL picture breaks down. On the high-temperature side \( T > T_{H} \approx J \), the exchange interactions are all negligible as we consider the thermodynamics. In this case, the Kubo-Tomita theory gives a constant linewidth \( \eta \sim J_{l}^{2}d_{L}^{2}/J \). On the low-temperature side \( T < T_{L} \), the two-leg ladder is in the gapped phase with \( \Delta_{a} \) and \( \Delta_{t} \) if the magnetic field is weak \( (H < \Delta_{a}) \). Thus, the linewidth decreases as \( T \searrow 0 \). The linewidth inevitably has an extremum around \( T = T_{L} \). From these arguments, the temperature dependence of the ESR linewidth in the whole temperature region is depicted as in Fig. 2.

One can similarly treat the effect of a transverse rung anisotropy \( H' = H'/r \sum_{j} S_{j}^{x} S_{j+1}^{x} \) with \( |\delta_{L}| \ll 1 \).

Table I. Orders of characteristic temperatures \( T_{L,H,cr} \) and the \( T \) dependence of the ESR linewidth \( \eta (T) \) in several quasi-1D AF systems. The temperature \( T_{H} \) \((T_{L}) \) is the higher (lower) boundary of the TLL phase. Constants \( A, B \) and \( C \) depend on the microscopic information on the systems. For comparison, we list the results of the \( S = 1/2 \) AF chains with XXZ anisotropy or an effective staggered field \( h \propto H \).

Here we show only the result: The contribution of the transverse rung anisotropy to the linewidth is a half of Eq. (10). It is also known that the linewidth of single spin chains with transverse anisotropy is a half of those with longitudinal anisotropy [6].

Next we move on to spin ladders with a leg anisotropy. We discuss a leg anisotropy \( H' = H'/r \sum_{j} S_{j}^{x} S_{j+1}^{x} \) where \( \delta_{L} \approx 1 \). The derivation of the linewidth is quite similar to the case of the rung anisotropy. We obtain the ESR linewidth

\[ \eta = \frac{2J_{l}^{2}}{\chi} \left( \frac{4C_{l}^{2}T}{\pi} + C_{l}^{4} \frac{T}{J} \right). \]

\[ \text{Note that the contribution from the staggered component is } T\text{-linear } \propto T/J \text{ in contrast to Eq. (10) because the interaction } (N_{z}^{2})^{2} \text{ is marginal as well as } J_{l}^{2}R_{L}^{2}. \] 

\[ \text{Thus, the exchange narrowing region is much wider than the case of the rung anisotropy. There is no reason to have an extremum around } T = T_{L} \text{ (Fig. 2(c)).} \]

Let us consider a case that the leg and rung anisotropies coexist. Since \( |J_{l}| \ll J \), the broadening \( \propto J_{l}^{2}T \) due to the leg anisotropy is masked by the large contribution \( \propto T \) of the leg anisotropy down to \( T \sim |J_{l}| \) if \( \delta_{L}(x) \) and \( \delta_{L}(y) \) are the same order. Namely, the crossover temperature \( T_{cr} \) is an order of \( |J_{l}| \) (Fig. 2(d)). From the above arguments, our prediction for spin ladders is summarized in Table I.

Generalizations. — Our theory on the two-leg ladder is immediately extended to other decoupled TLL phases of \( N \)-leg \( S = 1/2 \) spin ladders and tubes as long as the leg coupling \( J \) is much larger than rung couplings. Even if the system is composed of a complicated array of spin chains, \( A = [H', S'] \) is still involved only with couplings between two chains as long as interleg exchange interactions are short-ranged. The results of Fig. 2 and Table I are still valid in these cases. Moreover, one can apply our results to three-dimensionally (3D) coupled
spin chains as well. In this case, \( T_c \) is of order of an interchain coupling \( |J'| \) instead of \( |J| \) and the lower-temperature limit \( T_L \) is replaced to the 3D ordering temperature \( T \sim |J'| \) [36, 37].

According to Ref. [3], the linewidth is divergent at the Néel point \( T_N \). On the other hand, our theory predicts only the existence of the extremum at \( T_N \), which need not to diverge. In order to confirm whether the linewidth actually diverges or not, we have to carefully deal with 3D couplings beyond the random phase approximation [32].

We note that the growth of the linewidth with lowering temperature is similar to that of NMR relaxation rate [38, 39]. However, the growth of the ESR linewidth does not occur unless the rung anisotropy is absent, while that of the NMR relaxation rate always occurs regardless of anisotropies.

Comparison with Experiments. — We apply our results, Fig. 4 and Table I, to an \( S = 1/2 \) four-leg spin tube compound \( \text{CuCl}_2 \) [12] and an 3D weakly coupled \( S = 1/2 \) AF chains [29].

First we consider an \( S = 1/2 \) four-leg spin tube compound \( \text{Sul-Cu}_2\text{Cl}_4 \) [12]. Microscopic parameters of the model for this compound is yet to be settled [28, 11]. Nevertheless, since the \( T \) dependence of the susceptibility \( \chi \) for 50 K < \( T \) < 100 K is well reproduced by a bond alternating chain \( \mathcal{H} = J \sum_j (S_{2j} \cdot S_{2j+1} + \alpha S_{2j} \cdot S_{2j+1}) \) with \( J = 105.6 \) K and \( \alpha = 0.98 \) [23], we may conclude that the intraleg coupling \( J \) is much stronger than other interleg couplings. Figure 3 shows the experimentally derived linewidth [12]. Black curves in Fig. 3 are results of fittings with a function \( \eta = A/T + BT \) via constants \( A \) and \( B \) for several frequencies. The fittings with experimental data are excellent. From Fig. 3, we can extract the following information on \( \text{Sul-Cu}_2\text{Cl}_4 \). (i) The intraleg anisotropy \( \mathcal{H}_i \) is much weaker than the interleg one \( \mathcal{H}_o \) because the exchange narrowing region is not found. (ii) The temperature \( T_L \sim 20 \) K where the linewidth takes a maximum value is almost independent of the resonance frequency \( \omega \), namely, the applied magnetic field \( H \). This behavior shows a difference from the scenario of the staggered field [5].

Next we consider a classic \( S = 1/2 \) AF spin chain compound \( \text{CuCl}_2 \cdot 2\text{NC}_5\text{H}_5 \) [29]. The intrachain and 3D interchain couplings are evaluated as \( J = 13.4 \) K and \( J' = 0.12 \) K, respectively. The Néel ordered phase appears at \( T_N = 1.135 \) K due to \( J' \). Figure 4 shows the experimental ESR linewidth in this compound [29]. The data are fitted by a function \( \eta = A/T + BT \) via fitting parameters \( A \) and \( B \). In this compound, an energy scale \( T_c \sim 5 \) K exists, which means that the anisotropy comes from the interchain interaction. In the range \( T_c < T < T_N \sim 10 \) K, the exchange narrowing occurs, while for \( T_N < T < T_{cr} \), the linewidth rapidly increases obeying \( \eta \sim T^{-1} \) as \( T \) is lowered down to \( T_N \). Since nonuniversal factors \( C_{u,s} \) in Eqs. (10) and (11) are numerically determined for the spin chain [42, 43], further experiments with changing the direction of the magnetic field will enable one to determine strength of anisotropic exchange interactions of \( \text{CuCl}_2 \cdot 2\text{NC}_5\text{H}_5 \).

Conclusions. — In this paper, we have discussed a universal broadening of the ESR linewidth induced by weak interchain interactions in quasi-1D AF magnets. Our theory explains the observed broadenings in a four-leg spin tube compound (Fig. 3) and the high-temperature region above the 3D Néel ordered transition in a quasi-1D compound (Fig. 4). We emphasize that our theory is applicable to various spatially anisotropic quantum antiferromagnets. It will be interesting to apply our theory to spatially anisotropic frustrated quantum antiferromagnets. A more detailed analysis of the linewidth and the resonance frequency in weakly coupled AF chains will be published elsewhere [44].

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**BOSONIZATION FOR LADDERS**

In this section, we describe the non-Abelian and Abelian bosonization for the $S = 1/2$ two-leg spin ladder of Eq. (1). Suppose the interchain (rung) coupling is sufficiently weaker than the intrachain (leg) coupling, then we may start with two decoupled $S = 1/2$ Heisenberg Antiferromagnetic (AF) chains to analyze the ladder. The low-energy physics of a $S = 1/2$ single AF chain $\mathcal{H}_n$ is described by a gapless Tomonaga-Luttinger liquid (TLL). In the bosonization framework, the spin operator $S_{j,n}$ on the $n$th leg is written in two parts: $\mathcal{H}_n = \mathcal{H}_{\text{TLL}} + \mathcal{H}_{\text{fr}}$.

The uniform component $J_n = J_{nL} + J_{nR}$ is split into left-moving ($J_{nL}$) and right-moving ($J_{nR}$) parts. These fields are written in terms of chiral bosons $\phi_{nL(R)}$: 

$$J_{nL}^z = \frac{M}{2} + \frac{1}{\sqrt{2}} \partial_x \phi_{nL}, \quad J_{nR}^z = \frac{M}{2} + \frac{1}{\sqrt{2}} \partial_x \phi_{nR}, \quad J_{nL}^\pm = \frac{C_u}{2} e^{\pm i (\sqrt{\pi} \phi_{nL} + Hx/v)}, \quad J_{nR}^\pm = \frac{C_u}{2} e^{\mp i (\sqrt{\pi} \phi_{nR} + Hx/v)},$$

where $M$ is the magnetization per site, $\{S_{n,j}^z\}$ is split into two TLLs. The TLL Hamiltonian is expressed by using these fields as follows:

$$\mathcal{H}_n \simeq \int dx \frac{v}{2} [K(\partial_x \theta_n)^2 + K^{-1}(\partial_x \phi_n)^2],$$

where $v$ is the sound velocity of the TLL and $K$ is the TLL parameter. If the chain $\mathcal{H}_n$ has the spin-rotational SU(2) symmetry, the value of $K$ is fixed to unity, while magnetic anisotropies and the external field $H$ generally change the value. The staggered component $N_n$ is written by using an SU(2) matrix field $U_n$:

$$N_n = C_s \text{Tr}(U_n \sigma),$$

where $C_s$ is a dimensionless constant and $\sigma = (\sigma^x, \sigma^y, \sigma^z)$ is a set of Pauli matrices,

$$\sigma^x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma^y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma^z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

The element of the matrix field $U_n$ can be represented as

$$U_n = \frac{-i}{2} \begin{pmatrix} e^{i \sqrt{2} \pi \phi_n} & i e^{-i \sqrt{2} \pi \theta_n} \\ i e^{i \sqrt{2} \pi \theta_n} & e^{-i \sqrt{2} \pi \phi_n} \end{pmatrix}. \quad (S10)$$

Let us briefly discuss effects of the rung coupling and the excitation gaps of the ladder at zero magnetic field. Using the bosonization formula (S1), the rung interaction (3) in the paper is represented as

$$\mathcal{H}_{\text{rung}} = g_r \int dx \left( \cos \sqrt{4 \pi \theta_n} + \frac{1}{2} \cos \sqrt{4 \pi \phi_n} + \frac{1}{2} \cos \sqrt{4 \pi \phi_n} \right), \quad (S11)$$

where $g_r = J_r C_s^2$, and we have introduced new fields

$$\phi_{\pm} = (\phi_1 \pm \phi_2)/\sqrt{2}, \quad \theta_{\pm} = (\theta_1 \pm \theta_2)/\sqrt{2}. \quad (S12)$$

This rung interaction (S11) generates mass gaps of the $\phi_{\pm}$ sectors. According to Ref. 34, under zero magnetic field $H = 0$, two boson fields $\phi_{\pm}$ and their dual fields $\theta_{\pm}$ can be fermionized and the fermionized theory contains four Majorana fermions. Three of them have a degenerate excitation gap $\Delta_t = |g_r|/2$ and the other one has a larger gap $\Delta_s = 3|g_r|/2$. Physically, $\Delta_t$ is the spin-triplet (magnon) excitation gap and $\Delta_s$ is the spin-singlet (two-magnon bound state) gap.

**RETARDED GREEN’S FUNCTIONS**

In this section, we discuss a few important points in the calculation of the electron spin resonance (ESR) spectrum of the spin ladder (1). We consider the high-temperature region, $T \gtrsim \Delta_s$, in which the rung interaction $\mathcal{H}_{\text{rung}}$ and the perturbative anisotropy term $\mathcal{H}'$ are both negligible when we discuss the thermodynamic properties. In this region, the two-leg ladders are described by two TLLs:

$$\mathcal{H}_{\text{ladder}} \simeq \int dx \sum_{n=\pm} \frac{v}{2} \left( (\partial_x \theta_n)^2 + (\partial_x \phi_n)^2 \right). \quad (S13)$$

We should however note that when we consider the ESR spectrum of the ladder, we cannot neglect the effect of the magnetic anisotropy $\mathcal{H}'$ even in the high temperature region. This is because the ESR spectrum is essentially determined by the magnetically anisotropic terms even if they are very small.

Here we focus on the following longitudinal rung anisotropy:

$$\mathcal{H'} = J_r \delta_{\pm z} \sum_j S_{j,1}^z S_{j,2}^z. \quad (S14)$$
Following the standard bosonization techniques, the components of the operator $\mathcal{J}$, and $\mathcal{N}$, of Eqs. (6) and (7), can be expressed in terms of the boson fields as follows:

$$
\mathcal{J}_\perp = \frac{C_u}{\sqrt{4\pi}} \int dx \left[ \partial_x \phi_\perp (x) \epsilon^{-i\sqrt{4\pi}\phi_\perp + iHx/v} \cos \sqrt{4\pi} \phi_\perp - \partial_x \phi_\perp \epsilon^{-i\sqrt{4\pi}\phi_\perp + iHx/v} \cos \sqrt{4\pi} \phi_\perp \right]
$$

and

$$
\mathcal{N}_\perp = C_s^2 \int dx \left[ \sin(\sqrt{4\pi} \phi_\perp + Hx/v) \epsilon^{-i\sqrt{4\pi}\phi_\perp} \sin(\sqrt{4\pi} \phi_\perp + Hx/v) \right].
$$

Here new chiral boson fields $\phi_\perp(L,R)$ have been defined by

$$
\phi_\perp = \phi_\perp(L) + \phi_\perp(R), \quad \theta_\perp = \phi_\perp(L) - \phi_\perp(R).
$$

Following the standard bosonization techniques, the Fourier transformed retarded Green’s function of the operator $\mathcal{J}_\perp$ is calculated as

$$
G^R_{\mathcal{J}_\perp} (\omega) = \frac{NC_s^2}{\pi} G^R_{(1,1)} (\omega, H/v),
$$

where $N$ is the length of the leg and the Green’s function $G^R_{(1,1)} (\omega, H/v)$ is defined in Eq. (8). In order to obtain the ESR spectrum, we need $G^R_{\mathcal{J}_\perp} (\omega = H)$, namely, $G^R_{\mathcal{J}_\perp} (\omega, H/v)$. Instead of the general representation for $G^R_{\mathcal{J}_\perp} (\omega, q)$ of Eq. (8), we here comment on another useful representation,

$$
G^R_{\mathcal{J}_\perp} (\omega, q) = \frac{1}{\sin(\pi d)} \frac{1}{\Gamma(2d-1)} \left[ 2\pi T \right]^{2(d-1)}
$$

It is purely imaginary.

On the other hand, $G^R_{\mathcal{N}_\perp} (\omega)$ is proportional to $G^R_{\mathcal{J}_\perp} (\omega, H/v)$:

$$
G^R_{\mathcal{N}_\perp} (\omega) = \frac{NC_s^4}{4} G^R_{\mathcal{J}_\perp} (\omega, H/v).
$$

The right hand side is divergent if we take $\omega = H$. In fact, if we put $d = 1 + \Delta'$ with $|\Delta'| \ll 1$ in Eq. (S1), we obtain

$$
G^R_{\mathcal{J}_\perp} (\omega, H/v)
$$

Taking the limit $d \rightarrow 2$ in Eq. (S19), we obtain

$$
G^R_{(1,1)} (H, H/v) = \lim_{d \rightarrow 2} \frac{\sin(\pi d/2)}{\sin(\pi d)} \left( \frac{2\pi T}{v} \right)^{2} \times \left| \frac{1}{\Gamma(1 + i\frac{H}{2\pi T})^2} \sin \left( i \frac{H}{2T} \right) \right|
$$

It is purely imaginary.

where $\Delta'$ is approximately given by

$$
\Delta' \approx \frac{1}{\Delta} \left( \frac{\pi_2^2 T}{2\pi^2} \right)^{2}.
$$

Since $\mathcal{A}$ of Eq. (5) is proportional to the small parameter $\delta_\perp$, we did not need to carefully consider the effect of the renormalized TLL parameter on $\mathcal{J}_\perp$ and $\mathcal{N}_\perp$ in the main text. Namely, the renormalization effect leads to just a higher order correction to $\mathcal{A}$ and the ESR spectrum.

Secondly we find that the divergent term $\propto \Delta'^{-1}$ in Eq. (S21) cancels out the small factor $\delta_\perp^2$ in Eq. (9). As a whole, the physical quantity $G^R_{\mathcal{A}_{\Delta T}} (H)$ hence becomes finite. The point is that the divergent correlation $G^R_{\mathcal{J}_\perp} (H/v)$ always appears in physical quantities with a form $\delta_\perp^2 G^R_{\mathcal{J}_\perp} (H/v)$ so as to be finite.
Here, let us shortly consider effects of weak three-dimensional (3D) couplings between one-dimensional (1D) AF magnets (chains and ladders) on ESR. The 3D couplings generally modify the magnetic susceptibility from that of 1D quantum magnets, especially, in the low temperature regime of the TLL phase of the 1D systems. For purely 1D quantum spin chain systems, the susceptibility $\chi^{1D}$ depends on the frequency $\omega$ and the wavenumber $q_x$ along the chain direction: $\chi^{1D}(\omega, q_x)$. On the other hand, for 3D systems of coupled spin chains, the susceptibility $\chi^{3D}$ also depends on wavenumbers $(q_y, q_z)$ perpendicular to the chain direction: $\chi^{3D}(\omega, q_x, q_y, q_z)$. Within the random phase approximation (RPA) for interchain couplings, the susceptibility of 3D coupled spin chains is given by \[ \chi^{3D}(\omega, q_x, q_y, q_z) = \frac{\chi^{1D}(\omega, q_x)}{1 + F(q_y, q_z)\chi^{1D}(\omega, q_x)}. \] (S25)

The form of interchain couplings determines the function $F(q_y, q_z)$: For instance, $F(q_y, q_z) = J'(\cos q_y + \cos q_z)$ for spin chains embedded in the cubic lattice with an interchain coupling $J'$ between neighboring chains. The formula (S25) is valid above the 3D ordering temperature. The point is that the factor $F(q_y, q_z)$ is real. Let us assume $\chi^{1D}(\omega, 0) = A/(\omega - \omega_r + i\eta)$ near the ESR peak frequency (This is valid for TLL phases). Then, the 3D susceptibility at $(q_x, q_y, q_z) = (0, 0, 0)$ becomes

$$
\chi^{3D}(\omega, 0, 0, 0) = \frac{A}{\omega - \omega_r + 2AJ' + i\eta}.
$$

Within RPA, the three-dimensional coupling merely turns into an effective magnetic field which shifts the ESR frequency by $-2AJ'$. Therefore, in order to discuss effects of the 3D coupling on the ESR linewidth, we need to go beyond RPA.