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Direct determination of the Tomonaga-Luttinger parameter K in quasi-one-dimensional spin systems

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We test the analytical formula for the enhancement of the nuclear magnetic resonance rate T_1^{-1} by the critical spin fluctuations, over the simple power-law dependence predicted for a purely one-dimensional spin system, recently derived in the random phase approximation [M. Dupont *et al.*, Phys. Rev. B **98**, 094403 (2018)]. This prediction is experimentally confirmed by excellent fits to the published temperature dependence of T_1^{-1} data in the two representative spin compounds, $(C_7H_{10}N)_2CuBr_4$ (DIMPY) and $BaCo_2V_2O_8$, providing at the same time a *direct* and convenient experimental determination of the Tomonaga-Luttinger-liquid parameter K, very well in agreement with theoretical predictions.

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The Tomonaga-Luttinger liquid (TLL) theory provides a general low-energy description, including interactions, for any gapless one-dimensional (1D) system [1]. Its importance in the description of quasi-1D materials is therefore crucial, and it can be regarded as analogous to what the Fermi-liquid description is for three-dimensional (3D) systems. While the main hallmark of the TLL description, namely the power-law dependence of 1D response/correlation functions, had been experimentally well established previously [2,3], it is only a decade ago that quasi-1D quantum spin compounds have provided the final quantitative verification of the TLL theory [4,5]. In the spin-ladder compound $(C_5H_{12}N)_2CuBr_4$, also known as BPCB, one could compare the experimental values with the TLL-based predictions for the magnetic field (B) dependence of (i) the phase boundary $T_c(B)$ of the low-temperature (T) ordered phase, (ii) the low-T limit of the order parameter of this phase [4,6], and (iii) the nuclear magnetic resonance (NMR) spin-lattice relaxation rate $T_1^{-1}(B)$ in the TLL regime, at $T \gg T_c$ [4,7]. A successful theoretical description of these data thus confirmed the field-induced variations of the two TLL parameters: a dimensionless interaction parameter K that defines the power-law exponents and the renormalized Fermi velocity u. In these systems, B plays the role of the chemical potential controlling the filling of the (spinless) fermion band in the representation onto which the spin system can be mapped. The interaction between fermions depends on the filling of the band, which is notably reflected in the $K(B)$ dependence.

However, the first attempt to directly determine the K parameter from the measured $T_1^{-1}(T)$ dependence, performed in the spin-ladder compound $(C_7H_{10}N)_2CuBr_4$, also known as DIMPY, failed [8]. This was attributed to the enhancement of relaxation by the critical spin fluctuations in a very broad vicinity of T_c. Indeed, a correct determination of the K value from the power-law exponent of the $T_1^{-1}(T)$ temperature dependence is in practice precluded by the enhancement of relaxation related to the nearby T_c on the low-T side, as well as by the inherent limitation of a TLL description to low energy, and thus low temperature, on the high-T side [9,10]. Recently, this was described theoretically both by quantum Monte Carlo (QMC) numerical simulations, and analytically, using the random phase approximation (RPA) to describe the effect of fluctuations [11]. The former approach showed that a *purely* 1D (TLL) power-law regime of $T_{1\mathrm{TLL}}^{-1}(T) \propto T^{1/2K-1}$ dependence, observed when the three-dimensional (3D) exchange couplings are three or more orders of magnitude smaller than 1D coupling, rapidly shrinks and disappears as soon as the 3D coupling strength raises to the level of percent (see Fig. 4 in Ref. [11]). In practice, this means that it is not really expected to be observable in most of the experimentally interesting spin systems. Furthermore, a closed analytical expression, depending only on T_c and K, was derived within the RPA approximation (and checked against QMC) to take into account the fluctuations related to T_c [11].

Here, we apply this RPA correction to the published $T_1^{-1}(T)$ NMR data in two very different, representative, quasi-1D spin systems [8,12], and find that it provides a remarkable fit to the data. These fits present a *direct* experimental determination of the K values that confirms the theoretically predicted values. They also provide a convenient means of the experimental characterization of a quasi-1D system, independent of its complete theoretical description that requires the knowledge of the Hamiltonian and of numerical techniques [QMC, density-matrix renormalization group (DMRG)]. Finally, the fit covers the data quite close to T_c and can also provide an

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independent estimate of the T_c value. Altogether, it constitutes a reference for the normal quasi-1D behavior, which can be used to reveal nonstandard cases.

In the following, we first discuss the analytical RPA correction to the TLL prediction $T_1^{-1}(T)$, which was cast to a multiplicative correction function $\Phi(K, T_c/T)$ [11],

$$T_1^{-1}(T,B) = T_{1\mathrm{TLL}}^{-1}[T, K(B)] \times \Phi[K(B), T_c/T] = a\, T^{1/2K-1} \Phi(K, T_c/T), \quad (1)$$

where

$$\Phi(K, T_c/T) = \frac{1}{N(K)} \int_{-\infty}^{+\infty} \frac{d\xi}{\sin^2\left(\frac{\pi}{8K}\right) + \sinh^2(\pi\xi)} \times \left| \frac{\Gamma\left(\frac{1}{8K} + i\xi\right)}{\Gamma\left(1 - \frac{1}{8K} + i\xi\right)} \right|^2 \times \frac{\mathrm{E}\left[\left(\frac{T_c}{T}\right)^{4-1/K} \left| \frac{\Gamma\left(1-\frac{1}{8K}\right)\Gamma\left(\frac{1}{8K}+i\xi\right)}{\Gamma\left(\frac{1}{8K}\right)\Gamma\left(1-\frac{1}{8K}+i\xi\right)} \right|^4 \right]}{1 - \left(\frac{T_c}{T}\right)^{4-1/K} \left| \frac{\Gamma\left(1-\frac{1}{8K}\right)\Gamma\left(\frac{1}{8K}+i\xi\right)}{\Gamma\left(\frac{1}{8K}\right)\Gamma\left(1-\frac{1}{8K}+i\xi\right)} \right|^4} \quad (2)$$

and

$$N(K) = 2\Gamma^2\left(\frac{1}{4K}\right) \cos\left(\frac{\pi}{4K}\right) \mathrm{B}\left(\frac{1}{4K}, 1 - \frac{1}{2K}\right).$$

Here, $\mathrm{E}(x)$ is the complete elliptic integral of the second kind, $\Gamma(x)$ and $\mathrm{B}(x, y)$ are respectively the Euler gamma and beta functions, and a is the amplitude whose magnetic field dependence (not studied here) is determined from the complete expressions for the $T_{1\mathrm{TLL}}^{-1}(T, K)$ given in Refs. [4,5] (see also Ref. [13]). The correction Φ depends on K and T_c only, and not on other parameters of the system. Comparison to QMC results showed that this analytical "RPA + TLL" fit is expected to make the experimental determination of K possible even for weakly 1D spin systems, where the ratio of 3D to 1D couplings is as big as 10% [11].

A 3D plot of the $\Phi(K, T_c/T)$ function (Fig. 1) shows that the enhancement of relaxation is moderate, reaching a factor of 2 at temperature 10% above T_c. Its K dependence is quite feeble, as the contour lines only weakly bend away from the K-axis direction. At $T \gtrapprox 2T_c$ the enhancement falls below 5%, and can thus only weakly affect the *field* dependence of T_1^{-1} measured at fixed T well above T_c. This *a posteriori* justifies the use of a pure 1D $T_{1\mathrm{TLL}}^{-1}$ expression to approximately fit the measured field dependence of relaxation (typically recorded at twice the maximum T_c value) [4], also proposed to reveal the attractive ($K > 1$) or repulsive ($K < 1$) nature of a quasi-1D spin system [7].

Φ is normalized to 1, $\Phi(K, T_c/T \to 0) \to 1$, which ensures that the $T_1^{-1}(T)$ on increasing temperature converges to its TLL limit (Fig. 1). However, having a correction of about 5% at $2T_c$ means that the apparent power-law fit that neglects the RPA enhancement, taken e.g., in the temperature interval between $2T_c$ and $3T_c$, as in the previous analysis of DIMPY data [8], is significantly distorted. For the DIMPY data, this effect is quantified in Fig. 2: Indeed, the K values corresponding to the apparent power-law fit are systematically higher than the ones using the RPA + TLL fit defined by Eq. (2).

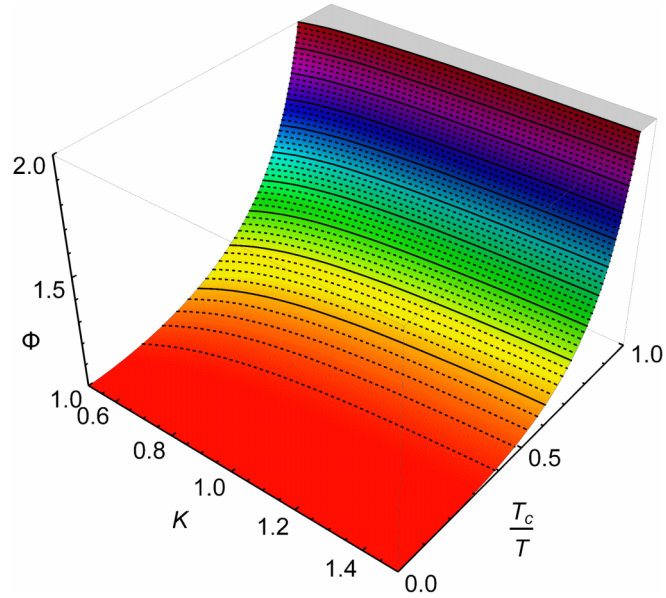

FIG. 1. The 3D plot of the correction function $\Phi(K, T_c/T)$ defined by Eq. (2) [11]. The dashed and solid contour lines are spaced at intervals of 0.02 and 0.1, respectively.

For these latter fits, T_c has been determined independently for each field value from the onset of building up of the order parameter, observed through the splitting of the NMR lines [see Fig. 1(b) in Ref. [8]]. Only two parameters are then determined by the least-squares fit to the data, the amplitude a and the K value. The K values obtained in this way nicely follow the theoretical prediction, with the exception of the data taken at 3.5 T. We remark that this lowest field value is relatively close to the critical field $B_{c1} = 2.9$ T, in the vicinity of which the TLL description is not applicable. Finally, the error bars of the RPA + TLL fits are smaller because the temperature interval of these fits is much wider (on the logarithmic scale), which stabilizes the fit.

While the $\Phi(K, T_c/T)$ function (Fig. 1) is weakly dependent on K, it clearly diverges as T decreases towards T_c [14]. Therefore, T_c can be taken as the third free parameter of the fit, in addition to K and a in Eq. (1), in order to provide an independent estimate of its value, predicted (extrapolated) from the spin dynamics observed above T_c. We present such three-parameter fits on the example of the published T_1^{-1} data in $BaCo_2V_2O_8$, an Ising-like $S = 1/2$ spin chain [12]. For these fits we note that the correction function $\Phi(K, T_c/T)$ has been calculated [11] for systems, such as Heisenberg $S = 1/2$ spin ladders, where the dominant spin fluctuations are the antiferromagnetic (AF) transverse ones, which is expected to be valid when $K > 0.5$. It is easy to show that it can also be applied to systems, such as Ising-like chains, where the dominant fluctuations are longitudinal and incommensurate (IC), which is expected to be valid when $K < 0.5$. The formulas that describe the relevant spin correlators and spin susceptibilities for these two types of fluctuations, given by Eqs. (6.47), (6.50), and (6.53) in Ref. [1], have an identical form up to the $1/2K \leftrightarrow 2K'$ symmetry transformation/correspondence around a so-called "η-inversion" point at $K = 0.5$ or $\eta = 1$ ($\eta = 1/2K$ [12,15,16]), at which the dominant fluctuations

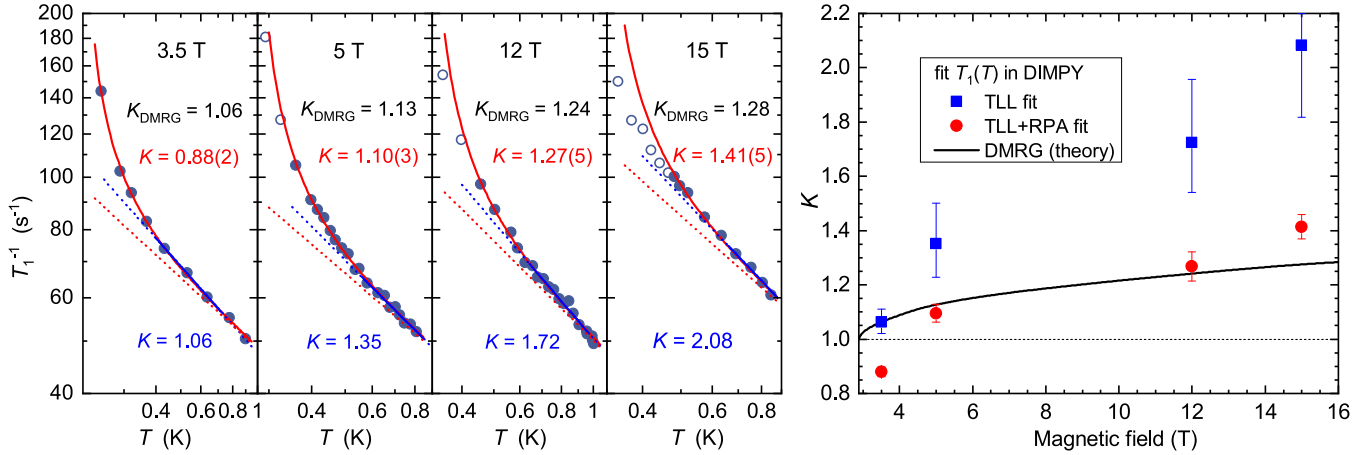

FIG. 2. Left panels: Comparison of the present RPA + TLL fit (red lines and text) with the previous apparent power-law (TLL) fit (blue lines and text) to the DIMPY data, as given in Ref. [8]. Solid data points denote values taken into account in the former least-squares fit. Pure uncorrected TLL contribution to this fit, $T_{1TLL}^{-1}(T,K) = a\,T^{1/2K-1}$, is given by red dotted lines, to show how much it differs from the apparent power-law fit. The temperature scale of each subpanel starts with the T_c value. K_{DMRG} values refer to the theoretical prediction obtained by DMRG in Ref. [8], whose field dependence is compared to the experimental K values in the right panel.

change their type. As both the RPA correction function and T_c are calculated/defined from the dynamic susceptibility, the same symmetry transformation applies to $\Phi(K, T_c/T)$. Therefore, for the longitudinal IC fluctuations we get

$$T_1^{-1}(T) \propto T^{2K-1}\Phi(1/4K, T_c/T). \tag{3}$$

Figure 3 shows this fit applied to the $BaCo_2V_2O_8$ data taken at 4.1 T [12], where both K and T_c (and the amplitude) are taken as the fit parameters. Here, the fitted data cover a broad enough temperature interval to well represent both the power-law and the fluctuations-enhanced regime. This is followed by a sharp peak of $T_1^{-1}(T)$, whose maximum reflects the maximum of the critical spin fluctuations and thus precisely defines the T_c value. The corresponding T_c value determined from the RPA + TLL fit given by Eq. (3) is only 2% lower, which is within the statistical error as defined by the fit. The equality of these two very different estimates of T_c, one reflecting critical dynamics *at* T_c and the other *above* T_c, constitutes a very strong confirmation for the validity of the employed correction function. Furthermore, the obtained $K = 0.23(1)$ value is very close to the $K = 1/4$ value expected for the nearby critical field value $B_c = 3.8$ T. Parenthetically, we observe that the $1/2K \leftrightarrow 2K'$ symmetry connects this value to the noninteracting limit $K' = 1$.

Figure 4 presents the fits to the two available $BaCo_2V_2O_8$ data sets close to the saturation field $B_s = 22.8$ T [12]. Here, the successful fit is of "mixed" character, $T_1^{-1}(T) \propto T^{2K-1}\Phi(K, T_c/T)$: The power-law (TLL) contribution is of

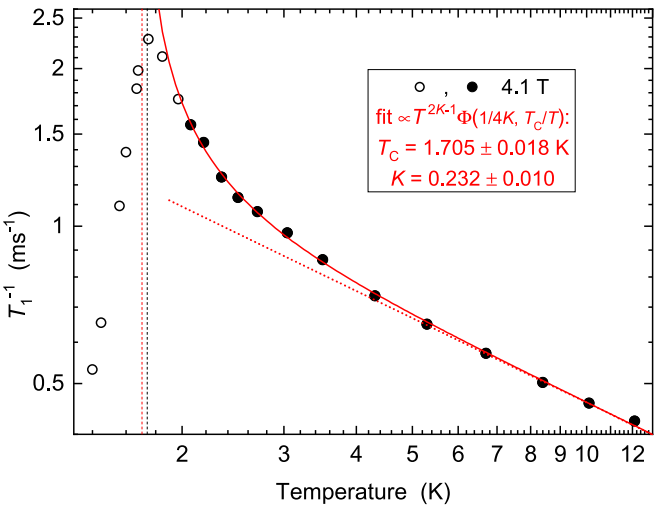

FIG. 3. The RPA + TLL fit (red solid line) of the $BaCo_2V_2O_8$ $T_1^{-1}(T)$ data (circles) recorded at 4.1 T [12], where solid circles denote the fitted points. The red dotted line is the pure T_{1TLL}^{-1} contribution to this fit. Vertical dashed lines denote the T_c determined by this fit (in red) and from the position of the $T_1^{-1}(T)$ maximum (in black), determined using the spline interpolation through the data points (not shown).

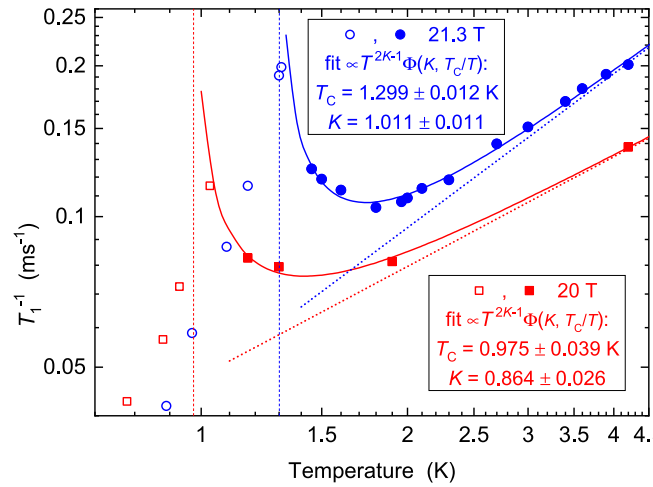

FIG. 4. The RPA + TLL fits (solid lines) of the $BaCo_2V_2O_8$ $T_1^{-1}(T)$ data [12] recorded at 21.3 T (blue color code) and 20 T (red color code), where solid symbols denote the fitted points. Dotted lines are the pure T_{1TLL}^{-1} contribution to this fit. Vertical dashed lines denote the T_c as determined by the fits.

the same type as at low fields (Fig. 3), corresponding to IC fluctuations, while the correction factor Φ corresponds to the dominant transverse AF fluctuations, as expected in the η-inversion scenario [16]. The power-law exponent can then be explained by the nature of the hyperfine coupling in this compound, which filters out the contribution of AF spin fluctuations and thus selects the IC contribution to T_1^{-1} [12], even when it is subdominant. As expected, the obtained K values are close to the $K = 1$ value that is predicted for the nearby saturation field, and they decrease with the field. Therefore, this fit supports the η-inversion scenario at high fields, as also suggested from the neutron data [17], but unlike the previously proposed interpretation of the NMR data, based on the pure TLL description [12]. However, for the present fit, it is not clear why the RPA correction factor of the subdominant fluctuations should be the same as for the dominant ones.

The above examples show that the RPA + TLL fit can be successfully applied to cover different types of fluctuations spanning the complete theoretical range of K values in various quasi-1D spin systems. The fit provides the K value that experimentally characterizes a quasi-1D spin system independently of the availability of a theoretical description. The latter can be unavailable because the microscopic Hamiltonian is only partially defined/known, e.g., when the phase diagram extends up to very high magnetic field values that are beyond the current experimental reach. The temperature range successfully covered by the fit typically goes quite close to T_c, down to about $1.2T_c$. This strong extension of the applicable range makes the fit more stable and possible even for systems farther away from the 1D limit. Below $\approx 1.2T_c$, we expect that the nature of the critical fluctuations changes from the 1D-based one, taken into account by Φ, to the usual 3D fluctuations, whose typical extension in temperature is of the order of 10%. Additionally, real compounds often present some sort of disorder, leading to a distribution of T_c values and the corresponding broadening of the peak of the measured $T_1^{-1}(T)$ data that reflect the critical fluctuations.

We remark that the $\Phi(K, T_c/T)$ function in principle depends on the geometry of 3D couplings, and that its analytical expression given by Eq. (2) has been calculated for the system of tetragonal symmetry [11] [see Supplemental Material (SM) for further details [18]]. We have also tested how its form is modified as a function of growing orthorhombic asymmetry (Fig. S1 in SM [18]). It turns out that this modification can be neglected up to approximately $J_x/J_y \simeq 2$, a point at which the asymmetry-induced enhancement of the function can be compensated by the effective/fictive increase of the fitted T_c by only 2.6%. In general, when both the geometry and size of the 3D couplings are known, and their q_z dependence is not frustrated, we can easily compute the *exact* corresponding Φ function [18]. However, for most of the real compounds the size of the 3D couplings is *not* known, and we can thus use Eq. (2) as a suitable proxy for systems that are *not* strongly anisotropic, and its generalization to the orthorhombic symmetry given by Eq. (S9) in SM [18] to describe other systems. Finally, in SM we also discuss how the $\Phi(K, T_c/T)$ function is evaluated and used in nonlinear fits to $T_1^{-1}(T)$ data in practice, and provide a simple example of the *WOLFRAM MATHEMATICA* code we used in our fits [18].

In conclusion, we performed a direct comparison between experimentally determined and theoretically predicted values of the parameter K that characterizes the power-law dependences predicted by the TLL description of quasi-1D systems. Using the recently proposed RPA-based correction factor that accounts for the enhancement of the NMR T_1^{-1} rate induced by critical fluctuations [11], we successfully fitted the observed $T_1^{-1}(T)$ dependence in two quasi-1D spin systems, DIMPY and $BaCo_2V_2O_8$, covering very different regimes of K values. This analysis establishes a simple reference procedure for the characterization of quasi-1D systems. It thus enables us to recognize such systems in compounds whose effective dimension is not evident/known. In particular, it provides a basis to distinguish between quasi-1D and quasi-2D spin systems, whose spin dynamics remains to be characterized. Finally, the RPA correction has been discussed here for the T_1^{-1} data, but it is expected to be relevant to other observables, such as, e.g., specific heat, for which its effect/size remains to be investigated.

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[18] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevB.101.220406 for the definition and the ways of calculating the $\Phi(K, T_c/T)$ function, as well as an example of the "RPA + TLL" fits.