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Wilson Ratio of a Tomonaga-Luttinger Liquid in a Spin-1/2 Heisenberg Ladder

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Using micromechanical force magnetometry, we have measured the magnetization of the strong-leg spin-1/2 ladder compound $(\text{C}_7\text{H}_{10}\text{N})_2\text{CuBr}_2$ at temperatures down to 45 mK. Low-temperature magnetic susceptibility as a function of field exhibits a maximum near the critical field H_c at which the magnon gap vanishes, as expected for a gapped one-dimensional antiferromagnet. Above H_c a clear minimum appears in the magnetization as a function of temperature as predicted by theory. In this field region, the susceptibility in conjunction with our specific heat data yields the Wilson ratio R_W . The result supports the relation $R_W = 4K$, where K is the Tomonaga-Luttinger-liquid parameter.

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One of the crucial parameters that characterize Fermi liquids, such as conduction electrons and liquid ^3He at low temperatures, is the Wilson ratio [1]

$$R_W = \frac{4}{3} \left(\frac{\pi k_B}{g\mu_B} \right)^2 \frac{\chi}{C/T}, \quad (1)$$

the dimensionless ratio of the temperature-independent magnetic susceptibility χ to the coefficient of the T -linear specific heat C . By dividing out the contribution of enhanced mass, which enters both χ and C , this parameter quantifies spin fluctuations that enhance the susceptibility. For this reason, it serves as a powerful tool to classify heavy fermion systems [2]. For instance, R_W is 2 for the $S=1/2$ Kondo lattice in the single-impurity limit [1] in contrast to 1 for non-interacting fermions.

In liquid ^3He , the archetypical Fermi liquid, R_W varies only weakly with pressure unlike the strongly pressure-dependent effective mass and is close to 4, the approximate limiting value for the Hubbard model with critical on-site repulsion [3]. This has promoted the view that quasiparticles in liquid ^3He are nearly localized [4].

In one dimension (1D), where an arbitrarily weak interaction renders the quasiparticle lifetime shorter than $\hbar/(E - E_F)$ because of stringent constraints the spatial dimension imposes on energy and momentum conservation in scattering processes, the Fermi liquid theory breaks down entirely, giving way to the Tomonaga-Luttinger liquid (TLL) as the correct low-energy description of fermions [5]. In a TLL, low-lying excitations are massless, collective bosonic modes instead of fermionic quasiparticles endowed with effective mass. Nonetheless, the TLL and the Fermi liquid are alike in that in both systems, χ is independent of temperature T and C is linear in T . As a result, the Wilson ratio remains a crucial

parameter in 1D—in fact even more crucial than in three dimensions, since a large variety of interacting systems in 1D fall into the TLL universality class and since each branch of bosonic modes in a given TLL is completely specified by just two parameters, the velocity v and the TLL parameter K [5, 6].

For instance, the Wilson ratio of 1D electrons contains only the velocities of spin and charge excitations, v_σ and v_ρ , and the TLL parameter $K_\sigma=1$ of the spin sector: $R_W = 2K_\sigma(1+v_\sigma/v_\rho)^{-1}$ [7]. In the non-interacting limit, where $v_\sigma=v_\rho=1$, R_W is again 1 as expected. With on-site Coulomb repulsion, v_σ decreases whereas v_ρ increases, resulting in R_W approaching 2 in the strongly repulsive limit [7, 8].

Another example of a TLL is a 1D antiferromagnet in a gapless regime—such as a spin-1 linear chain and a spin-1/2 ladder, both in magnetic fields larger than the magnon gap, or a spin-1/2 linear chain in zero field as well as in magnetic fields [6]. There, $\chi = (g\mu_B)^2 K/(\pi v)$ and $C = \pi k_B^2 T/(3v)$, and thus the Wilson ratio must obey the relation

$$R_W = 4K, \quad (2)$$

which to our knowledge has not been noted before [9]. It is remarkable that except for the trivial numerical factor, the Wilson ratio is the TLL parameter K , which governs the exponents of all spin correlation functions, including dynamic ones. This is not surprising, however, since TLLs are quantum critical [10] and, consequently, their dynamic properties are inextricably linked to static properties [11].

Despite the significance illustrated by these examples, the Wilson ratio has never been determined experimentally in a TLL because of the lack of a good

material. Here we report the Wilson ratio of a strong-leg $S=1/2$ Heisenberg spin-ladder antiferromagnet in magnetic fields, determined from accurate micromechanical force magnetometry and specific-heat measurements. The result is in agreement with a density-matrix renormalization-group calculation in conjunction with Eq. 2.

For these measurements, we have chosen $(\text{C}_7\text{H}_{10}\text{N})_2\text{CuBr}_2$, known as DIMPY, in which Cu^{2+} spins form two-leg ladders along the $\mathbf{a}$ axis of the monoclinic lattice [12–16]. The ratio $x = J_{\text{leg}}/J_{\text{rung}}$ between the leg exchange and the rung exchange is 2.2(2) in this material, placing it in the rare, strong-leg regime. The zero-field magnetic excitations measured by inelastic neutron scattering are strongly dispersive along the ladder direction but dispersionless, within the instrumental resolution, along orthogonal directions. Specific heat shows no long-range magnetic order down to 150 mK [13] demonstrating that DIMPY is an excellent 1D system with a gapless, TLL phase above the critical field $H_c = 3.0(3)$ T, at which the magnon gap vanishes.

Magnetization measurements were performed on a $13.8\text{ }\mu\text{g}$ single crystal with 67% deuteration using a micromechanical Faraday balance [17]. The device was loaded into a dilution refrigerator with the crystallographic $\mathbf{c}$ axis of the sample parallel to the magnetic field. A separate coil provided a field gradient of 2 Tm^{-1} , which exerted the force $\vec{F} = \vec{M} \cdot \nabla \vec{H}$ on the sample, where $\vec{M}$ is the magnetization. The device is designed to directly measure this force, with negligible contributions from magnetic torque. For a given field gradient, the output of the device is directly proportional to M . The proportionality constant was obtained by comparing, at 1.8 K and 4.3 K, the device outputs with the magnetization curves of a 30.5 mg single crystal measured in a SQUID magnetometer.

Figure 1(a) shows the magnetization of DIMPY as a function of magnetic field. At 300 mK the magnetization is nearly zero at low fields except for an evident contribution of paramagnetic impurities, whose concentration is about 0.9%. Upon reaching the critical field H_c , at which the magnon gap closes, the magnetization increases rapidly [14]. This signature of H_c becomes less prominent as the temperature is raised. At 4.3 K, higher than the magnon gap $\Delta = 3.7(1)$ K [13], the magnetization curve is featureless, with a roughly constant rate of increase throughout the field range of our experiment.

Differentiating the magnetization with respect to the magnetic field yields the magnetic susceptibility as a function of the field, as shown in Fig. 1(b). The susceptibility is temperature-independent within our resolution at least up to 300 mK. In a truly 1D gapped antiferromagnet, the zero-temperature magnetic susceptibility exhibits a square-root divergence at H_c , a singularity characteristic of free 1D fermions to which the system can be

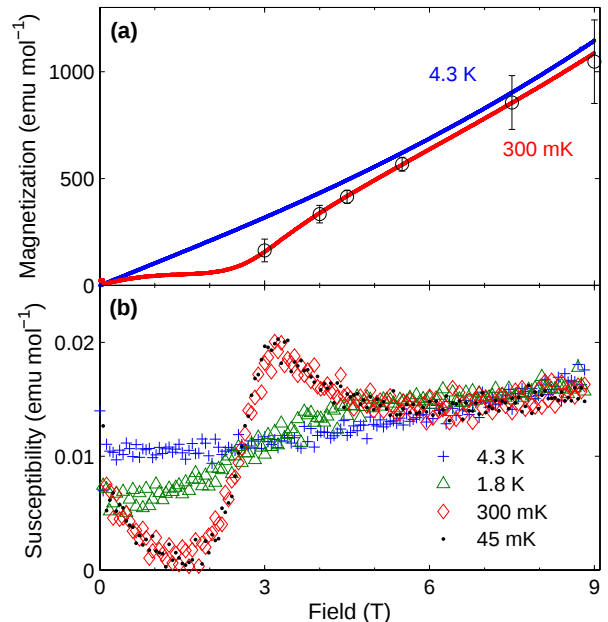


FIG. 1. (Color online). (a) Magnetization of DIMPY as a function of magnetic field at 300 mK and 4.3 K. The open circles represent DMRG results for $x = 2.0$ and $g = 2.2$. (b) Magnetic susceptibility as a function of magnetic field.

mapped. As the temperature rises, this singularity becomes a rounded peak near H_c [18]. Our low-temperature data clearly exhibit such a peak, providing strong evidence for the excellent 1D character of DIMPY in support of heat-capacity and inelastic neutron-scattering results [13].

Another characteristic feature of a gapped 1D antiferromagnet is a local minimum in the magnetization at temperature T_m , a minimum that marks the upper limit of the TLL temperature regime [18–20]. Such minima have been observed in the $S=1$ linear-chain compound NDMAP [21] and inferred for the $S=1/2$ strong-rung ladder material BPCB [22], but not in any other 1D antiferromagnets because of material problems such as the presence of field-induced staggered fields and too high a critical field for the present technology. To further test the prediction of Refs. [18–20] and, at the same time, to ensure that our low-temperature susceptibility data come from the TLL regime, we have measured the magnetization as a function of temperature for different magnetic fields $H \geq H_c$, as shown in Figs. 2(a) and (b). At 4 T, slightly above H_c , the magnetization reaches a minimum at about 0.7 K. The minimum at higher fields occurs at even higher temperatures, assuring that our susceptibility data at 45 mK and 300 mK are indeed from deep in the TLL regime, at least at and above 4 T.

Figure 2(c) presents T_m from the data as a function of

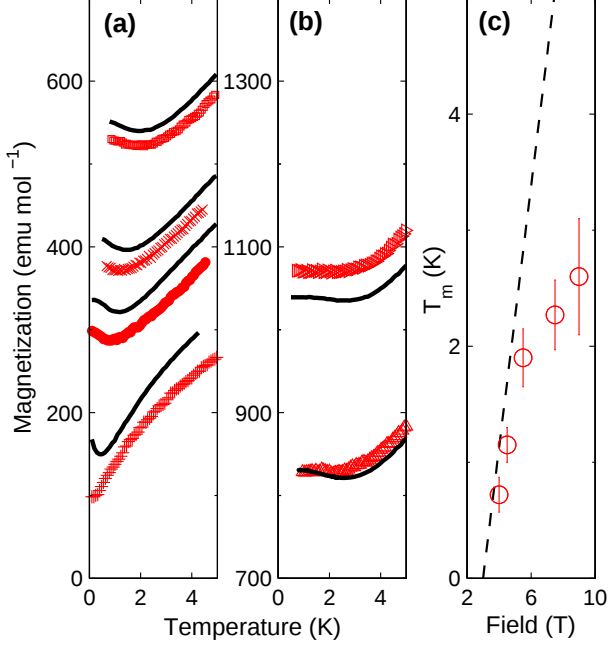


FIG. 2. (Color online). (a) Magnetization of DIMPY as a function of temperature. Symbols are experimental data, from which the magnetization of paramagnetic impurities has been subtracted; solid lines are quantum Monte Carlo simulations. Fields are, from bottom to top: 3 T, 4 T, 4.5 T, 5.5 T. (b) From bottom to top: 7.5 T, 9 T. (c) Position of the magnetization minimum from the data for $H \geq 4$ T as a function of magnetic field. Dashed line is the universal behavior, Eq. 3, for free fermions.

magnetic field, along with the universal relation

$$T_m = 0.76238 \frac{g\mu_B}{k_B} (H - H_c) \quad (3)$$

predicted by the free fermion theory with no adjustable parameters [20]. At fields near H_c , where the density of fermions to which the ground-state magnons are mapped is small, the theory and experiment are in excellent agreement. As the field increases, T_m falls below the universal relation, as predicted by QMC simulations [20]. This downward deviation is caused by repulsion between the fermions.

For further comparison with the experiment, we have performed QMC simulations of the magnetization of an $S = 1/2$ ladder consisting of 120 rungs, and have found that the g factor $g = 2.2$ and the exchange ratio $x = 2.0$, which is consistent with 2.2(2) mentioned earlier, give the best overall agreement with the data, as shown in Figs. 2(a) and (b). At each temperature and magnetic field, 50 runs of 1.2×10^7 Monte Carlo steps were used for averaging. The simulations and the experimental data share two important features. First, the magnetization

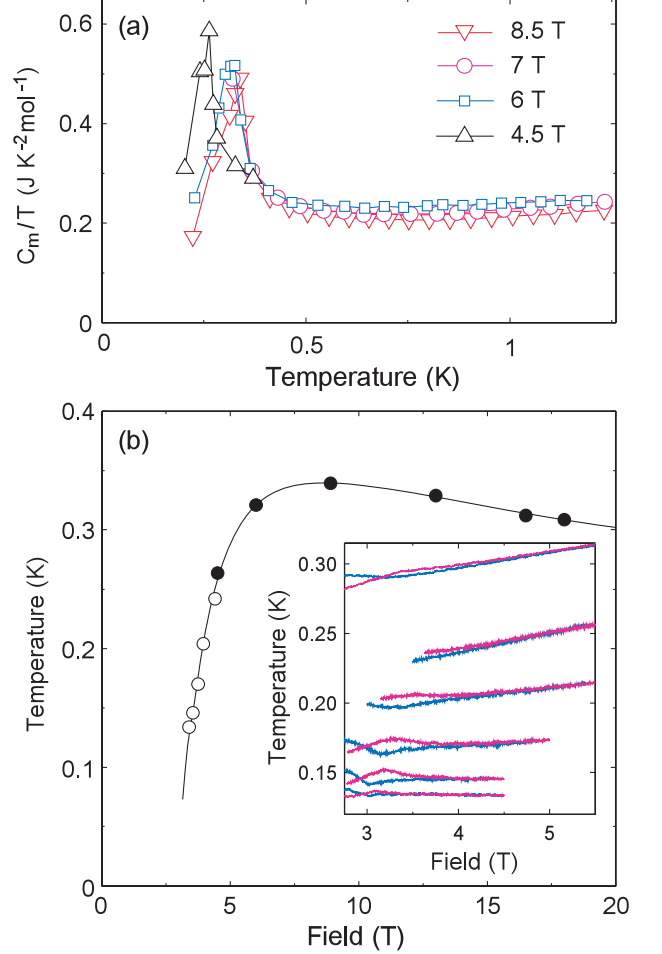


FIG. 3. (Color online). (a) Magnetic specific heat, C_m , of fully deuterated DIMPY, shown as C_m/T . (b) Ordering temperature as a function of magnetic field. Solid circles are from specific-heat peaks. Open circles are from the magnetocaloric-effect data shown in the inset, where pink and blue lines represent the sample temperature during upward and downward field sweeps, respectively. Other lines are guides to the eye.

minima appear at comparable temperatures, except at 3 T which is either at, or very close to, H_c . Second, as the magnetic field increases, the minimum becomes shallower. Moreover, density-matrix renormalization-group (DMRG) calculations with $g = 2.2$ and $x = 2.0$ —the values found by the QMC simulations—yield zero-temperature magnetization in excellent agreement with the 300 mK data, as shown in Fig. 1(a).

With the excellent one-dimensionality of DIMPY firmly established, we proceed to determine the Wilson ratio R_W . For this purpose, we have measured the specific heat of a 6.6 mg single crystal of fully deuterated DIMPY [23] using relaxation calorimetry [24], with the magnetic field applied along the c axis as in the magnetization measurements. Phonon, nuclear-spin, and

nuclear-quadrupole contributions to the specific heat were subtracted as described in Ref. [13] to obtain the magnetic specific heat C_m . At 6 T, 7 T, and 8.9 T, C_m exhibits T -linear behavior, demonstrated by constant C_m/T , at temperatures below about 1 K, except for a sharp peak signalling ordering at a lower temperature, as shown in Fig. 3(a). To detect ordering at magnetic fields close to H_c , we have also made magnetocaloric-effect measurements in the same calorimeter with the same sample, as shown in the inset to Fig. 3(b). From the specific-heat peaks and the magnetocaloric effect, the boundary of the ordered phase has been mapped to 18 T, as shown in Fig. 3(b). The phase boundary is highly asymmetric, peaking at about 8.9 T, where the fermion velocity is at or near maximum [13].

Ordering found in this sample at temperatures up to 340 mK is in strong contrast to the absence of ordering in a 67% deuterated sample at temperatures down to 150 mK [13], indicating that full deuteration in DIMPY dramatically enhances interladder exchanges. To further examine the effect of full deuteration, we have measured the magnetization of the sample as a function of the magnetic field at 1.8 K and 4.3 K in the SQUID magnetometer. The results were identical to those of the 67% deuterated sample within 2.0%, ensuring that the intra-ladder exchanges J_{leg} and J_{rung} —which govern the TLL physics of DIMPY—are unaffected by full deuteration.

C_m/T needed to determine R_W is obtained from the data at 6 T, 7 T, and 8.9 T at temperatures below 0.75 K, excluding the region affected by the peak. At 5 T and 8 T, C_m/T is taken from published data for a 67% deuterated sample [13]. For consistency, we use the susceptibility data taken at 300 mK rather than 45 mK, although the two are nearly identical. On assumption that $g=2.2$ in Eq. 1, R_W determined from the susceptibility and specific heat is shown in Fig. 4 as a function of the normalized magnetization $m = M(N_A S g \mu_B)^{-1}$. It takes on values close to 4, increasing with increasing m .

To examine whether this result validates Eq. 2, we have employed the DMRG method to calculate the TLL parameter K for $x=2.0$ [25]. The result, shown in Fig. 4 as R_W by assuming Eq. 2, is in good agreement with an earlier calculation [26]. Starting from 1 at $m=0$, K deviates upward from this universal free-fermion value—characteristic behavior of a strong-leg ladder [26] as opposed to a strong-rung ladder. R_W from the experiment on the other hand is not always larger than 4 as expected from Eq. 2. A plausible explanation is that this quantity is sensitive to the value of the g factor that enters Eq. 1. In fact, $g=2.04$ suggested by saturation-magnetization data [14]—instead of $g=2.2$ chosen here on the basis of the comparison of the magnetization data with the QMC simulations—will raise R_W by about 16%, bringing it to closer agreement with $4K$ from the DMRG calculations. Taking this into account, we conclude that within the combined uncertainties of experiment and calculations,

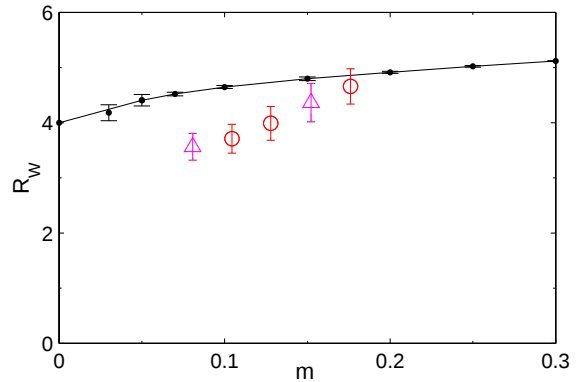


FIG. 4. (Color online). Dependence of the Wilson ratio R_W of DIMPY on the normalized magnetization m . Open symbols are experimental data, assuming $g=2.2$ in Eq. 1; specific-heat data from Fig. 3(a) have been used for circles, and those from Ref. [13] for triangles. The error bars represent only the combined uncertainties of the magnetization and heat capacity measurements; the uncertainty of the g factor has not been included. Small filled circles are the TLL parameter K for $x=2.0$, computed by the DMRG method and displayed as R_W by assuming Eq. 2. The line is a guide to the eye.

the result strongly supports Eq. 2, the equivalence between R_W and K .

To summarize, our magnetization measurements further confirm that DIMPY is an ideal 1D system whose gapless phase above H_c is a Tomonaga-Luttinger liquid (TLL) at low temperatures. The magnetization in this phase exhibits a minimum at T_m , whose dependence on magnetic field is in good agreement with quantum Monte Carlo simulations. The maximum in the low temperature susceptibility near H_c is consistent with the square-root singularity expected for gapped 1D antiferromagnets at zero temperature. The successful determination of the Wilson ratio, in conjunction with the density-matrix renormalization-group calculations of the TLL parameter K , makes DIMPY the first laboratory 1D system that lends support to the relation $R_W = 4K$.

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