Inhomogeneous ordering in weakly coupled Heisenberg $S = \frac{1}{2}$ chains with random bonds

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Long-range magnetic ordering in the quasi-one-dimensional random-bond antiferromagnet $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$ is studied in muon spin rotation experiments as a function of disorder strength. Compared to the disorder-free parent materials, the saturation-ordered moment is found to be considerably reduced. Moreover, even in weakly disordered species, the magnetically ordered state is shown to be highly inhomogeneous. The results are interpreted in terms of weakly coupled random spin chains, governed by the “infinite randomness” fixed point.

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I. INTRODUCTION

Arbitrary weak disorder can qualitatively alter one-dimensional spin systems. A case in point is the Heisenberg $S = 1/2$ antiferromagnetic chain with random bond strengths. Even though all exchange constants remain antiferromagnetic, the ground-state and low-energy excitations are quite distinct from those in Bethe’s uniform spin chain [1]. Regardless of the details of disorder, the system flows to the “infinite randomness” fixed point, represented by the so-called random singlet (RS) phase [2–5]. In the latter, all strong bonds are eliminated via a decimation procedure. What remains is a collection of noninteracting *effective* $S = 1/2$ antiferromagnetic dimers. The distribution of dimer strengths diverges at $\omega \rightarrow 0$ and is universal in this limit. As a result, all low-temperature thermodynamic properties and correlation functions acquire *universal* scaling forms.

Although the RS state is by now very well understood theoretically, experimental realizations are few. The first was quinolinium(TCNQ)₂ where the pioneering work of Tippie and Clark reported signs of RS physics in bulk properties [6]. The only other potential candidate is $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$ [7–10]. The disorder-free compounds with $x = 0$ and $x = 1$ are well characterized $S = 1/2$ chain systems [11–15]. The crystal structure is visualized in Fig. 1 [16]. The ($S = 1/2$)-carrying Cu^{2+} are bridged by superexchange interactions via O^{2-} ions, forming chains along the c axis of the orthorhombic structure. The corresponding antiferromagnetic exchange constants are $J = 280$ and $J = 540$ K for $x = 0$ and $x = 1$, respectively [11]. For the disordered derivatives $0 < x < 1$, the *average* values of J changes almost linearly with x [17]. However, due to the difference in the ionic radii of Si and Ge, the exchange constants are randomly distributed in the chains. A recent comparison of bulk measurements with numerical simulations has convincingly demonstrated that the distribution of J ’s is, to a good approximation, a simple bimodal one. In fact,

it is just a spatially random distribution of the values in the two parent compounds [9,10]. The randomness of the distribution reveals itself in an unusual behavior of the low-temperature magnetic susceptibility [7,9,17]. Unfortunately, an early inelastic-neutron-scattering confirmation of RS physics in $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$ [7] was later shown to be erroneous [8,18]. However, recent detailed macroscopic and NMR studies [9,10] as well as a theoretical paper [19] have firmly confirmed that for $x = 0.5$, $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$ indeed demonstrated all the key features of the RS state.

Due to weak interchain interactions, all $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$ materials order magnetically with Néel temperatures ranging from 1 to 10 K [11,14,17]. Therefore, all previous work aiming to probe RS physics was focused on the paramagnetic phase to avoid the effects of three-dimensional order. In contrast, the present paper focuses on the low-temperature ordered phase. We use a range of muon spin rotation (μSR) experiments to demonstrate that even in weakly disordered samples, the three-dimensionally ordered state is highly unusual. Not only is the ordered moment considerably reduced compared to the parent compounds, but also it is highly inhomogeneous. We argue that this inhomogeneity is not a simple reflection of the weak structural disorder but represents the intrinsic infinitely random nature of the RS state in individual chains.

II. EXPERIMENT

Single-crystal samples of $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$ with $x = 0, 0.05, 0.15, 0.3, 0.5, 0.95$, and 1 were grown using the floating-zone method. Some of the crystals were from the batch already used in previous studies [7,8,11–15,18]. Crystallographic characterization structure was performed on a Bruker AXS single-crystal diffractometer with an APEX II CCD detector with over 3000 Bragg intensities collected for each sample. Calorimetric measurements were performed with a Quantum Design physical property measurement system in the temperature range of 50 mK–250 K. These data were collected on small fragments of masses ranging between 7.5 and 22.4 mg. The main focus of this paper is on the μSR

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FIG. 1. (Color online) Schematic of the crystal structure of $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$, emphasizing the Cu-O spin chains.

experiments. These were performed on the General Purpose Spectrometer (GPS), Dolly, and Low-Temperature Facility (LTF) spectrometers at the Swiss Muon Source at the Paul Scherrer Institut. The samples were powders, obtained from parts of the as-grown single crystals. The samples were glued with General Electric varnish on a silver plate for the LTF or wrapped in silver polyester foil for the Dolly and GPS experiments. The μSR data were collected in a zero field (ZF), in weak transversal fields of 3 or 5 mT, and various longitudinal fields. The spectra were analyzed using MUSRFIT [20], a tool to analyze time-differential μSR data.

III. RESULTS AND DATA ANALYSIS

A. Crystallographic characterization

In all samples, single-crystal x-ray diffraction at room temperature is consistent with the orthorhombic space group $Pnma$. The measured x dependence of the lattice parameters in $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$ is monotonic and linear (Fig. 2, bottom), in agreement with previous studies [16,17]. A structural refinement confirmed that the actual Ge content in our samples is in excellent agreement with the nominal one (Fig. 2, top). Moreover, small samples taken from different parts of the as-grown crystalline rods showed no detectable composition variation.

B. Specific heat

With the main subject of this study being magnetic ordering in $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$, specific heat measurements were used as the most straightforward indicator of the corresponding phase transitions. The focus of these measurements was on Si-rich samples with $x \leq 0.5$. The magnetic contribution C_{mag} was estimated by subtracting a T^3 Debye lattice term, extrapolated down to low temperature from the range between 14 and 25 K. As shown in Fig. 3, top panel, a pronounced λ anomaly is observed in all samples studied. The feature remains sharp and well defined across the entire concentration range. Its position was associated with the Néel temperature T_N and is tabulated in Table I and plotted against concentration in Fig. 4 (right axis). The two end materials show the maximal T_N , whereas the lowest value is observed for $x = 0.5$. Note that our data on well-characterized single crystals are in contradiction with an earlier paper on polycrystalline samples [17]. In the

FIG. 2. (Color online) Top: Ge concentration determined from single-crystal x-ray diffraction for all samples as a function of nominal concentration. Bottom: Lattice parameters as a function of nominal concentration.

latter, two transitions were claimed for the low-concentration regime, and the magnitude of the λ anomaly was found to substantially decrease with increasing disorder. We suspect that the discrepancy is due to sample inhomogeneities in the polycrystal experiments.

A striking feature of the measured temperature dependencies of the specific heat is the change in the low-temperature asymptotic in disordered samples as compared to that in the parent material. To quantify this behavior, the data in the ranges of $0.25 \text{ K} < T < 0.7T_N$ ($x = 0.5$, $x = 0.3$), $0.65 \text{ K} < T < 0.7T_N$ ($x = 0.15$), and $1.9 \text{ K} < T < 0.7T_N$ ($x = 0$ and 0.05) were fit to a power law $C \propto T^\alpha$ (lines in the top panel of Fig. 3). The lower bound was imposed to avoid a Shottky-like contribution seen in all samples at the lowest temperatures, possibly due to nuclear spins [21]. The fitted exponent is plotted against Ge concentration in the bottom panel of Fig. 3. We see that with increasing x , the exponent rapidly crosses over from $\alpha \sim 4.4$ in the parent material to $\alpha \sim 1.8$ in the disordered compounds. Both these limiting values are distinct from expectations for a conventional antiferromagnet. The latter has a linear spin wave dispersion. The result is a density of spin wave states which is quadratic with energy and a Debye-like contribution to specific heat with $\alpha = 3$. The observed large value of α in the disorder-free material is most likely related to the small anisotropy gap previously observed in these compounds [13]. In contrast, a reduction in α with the introduction of disorder clearly indicates a proliferation of low-energy states compared to the simplest spin wave model.

FIG. 3. (Color online) Top: Measured temperature dependence of the specific heat C/T of $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$ for different concentrations (symbols). Solid lines are power-law fits to the data in the ordered state as described in the text. Bottom: composition dependence of the specific heat power law $C(T) \propto T^\alpha$.

C. μSR

1. Weak transversal field experiments

The μSR technique is used to accurately determine the magnetic volume fraction and the ordering temperature. The volume fraction is proportional to the initial asymmetry measured from the μSR spectra [22]. For five of our Si-rich samples the measured temperature dependence of the asymmetry is plotted in Fig. 5. Magnetic ordering is marked by a sharp steplike increase. At elevated temperatures, the sample is completely in the paramagnetic phase. The observed variation in asymmetry in this regime is an instrument effect.

TABLE I. Ordering temperature determined by specific heat and μSR measurements and apparent asymmetry step widths in transversal field experiments.

x	T_N (K) from $C(T)$	T_N (K) from μSR	ΔT (K)
0	8.9(1)	9.23(1)	0.163(10)
0.05	5.4(1)	5.68(1)	0.139(4)
0.15	2.94(6)	2.97(1)	0.057(3)
0.3	1.63(4)	1.65(1)	0.038(3)
0.5	0.98(4)		
0.95	4.0(1)	4.09(1)	0.108(11)
1	8.7(1)		

FIG. 4. (Color online) Concentration dependence of the ordering temperature T_N determined by specific heat and μSR measurements and saturation moment m_0 .

For low temperatures, our results indicate that all samples are entirely ordered. For $x \leq 0.15$ this is immediately apparent in the data collected on the Dolly and GPS instruments: At low temperatures the asymmetry is almost zero. For $x = 0.3$ we observe some residual asymmetry. This is, however, due to the about 20% background in the LTF instrument where these data were taken.

To determine the ordering temperatures, the data were analyzed using an empirical sigmoidal function,

$$A = A_2 + \frac{A_1 - A_2}{1 + \exp\left(\frac{T - T_N}{\Delta T}\right)}. \quad (1)$$

Here A_1 , A_2 are the asymmetry below and above T_N , respectively, and ΔT is the apparent step width. Note that nonzero step widths are intrinsic to the technique. On the other hand, a particularly broad step would indicate a distribution of

FIG. 5. (Color online) Measured temperature dependence of the asymmetry in a weak transversal field, which is proportional to the magnetic volume fraction, for five different concentrations. The solid lines are fits to the data to determine the ordering temperature T_N and transition width.

transition temperatures. The results of the fits are summarized in Table I and are plotted in Fig. 4. The excellent agreement with the calorimetric measurements is apparent. The step width is quite narrow and does not show any increase when x deviates from zero or unity. All this testifies to the quality and homogeneity of our samples.

2. Zero-field μ SR

The magnitude of the ordered magnetic moments was studied in zero-field μ SR experiments. Typical μ SR spectra collected in a series of $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$ samples at the lowest accessible temperature are shown in Fig. 6. Static internal fields due to magnetic order produce clear asymmetry oscillations with at least two different frequencies in the end compounds. The component of the internal field that is parallel to the incoming muon spin causes a finite nonoscillating asymmetry at late times (“tail”). The immediate effect of disorder, *even at very low concentration*, is the suppression of oscillations. The $x = 0.05$ and $x = 0.95$ materials still show at least two frequencies, but these are already heavily damped. At higher concentrations the spectra change to a

more Gaussian-like shape with the relaxation times decreasing towards $x = 0.5$.

For a quantitative analysis, let us first focus on the regimes $x \geq 0.95$ and $x \leq 0.05$ where damped oscillations are visible. The pure material is known to order in a commensurate nearly collinear antiferromagnetic structure [11]. Thus, the multitude of observed precession frequencies is due to different muon stopping sites. An adequate description of the data can be obtained assuming three distinct sites. For this model, the asymmetry as a function of time is empirically given by [22]

$$A = \frac{2}{3}[A_1 \cos(\omega_1 t + \phi) \exp(-\lambda_1 t) + A_2 \cos(\omega_2 t + \phi) \exp(-\lambda_2 t) + A_3 \cos(\omega_3 t + \phi) \exp(-\lambda_3 t)] + \frac{1}{3}(A_1 + A_2 + A_3) \exp(-\lambda_{\text{tail}} t) + A_{\text{bg}} \exp[-(\lambda_{\text{bg}} t)^\beta]. \quad (2)$$

The parameters A_1 , A_2 , and A_3 represent the muon fractions stopped at the different muon sites (site populations), respectively. Their ratio is not temperature dependent and was fit globally in each end compound. For the disordered samples, the ratio was assumed to be the same as in the corresponding parent compounds. In Eq. (2), A_{bg} is the fraction of the muons which stop in a paramagnetic part of the sample or possibly in the sample holder. Its value tends to be very small below T_N . The corresponding parameters λ_{bg} and β were fit to spectra above T_N and were kept fixed at low temperatures. λ_{tail} reflects the spin fluctuations of the muons with a polarization component parallel to the local magnetic field. We found it to be negligibly small and temperature independent. This implies that critical fluctuations are faster or slower than the accessible μ SR time window.

The parameter ϕ tended to 0 in the initial fits and was later fixed to $\phi = 0$ as appropriate for a simple commensurate antiferromagnetic structure. The parameters ω_1 , ω_2 , and ω_3 are the mean-precession frequencies at the three different muon sites in our model. They are related to the local internal magnetic fields on sites i through $B_i = \gamma_\mu \omega_i$ with $\gamma_\mu = 85.2 \times 10^{-3} \text{ rad s}^{-1} \text{ G}^{-1}$. The widths of the corresponding field distributions are described by the relaxation rates λ_1 , λ_2 , and λ_3 . The latter were found to vary only slightly below T_N . Upon introduction of disorder the relaxation rates increase considerably. Assuming that all the internal fields are proportional to the static ordered magnetic moment, the ratios $B_i:B_j$ were treated as temperature-independent parameters in each sample. The temperature dependence of the three internal fields for $x = 0$, 0.05 , 0.95 , and 1 are shown in Figs. 7(a)–7(d). They show typical order-parameter behavior. Within each sample, the saturation fields at the three sites are rather different. Their ratios are concentration dependent. Figure 7(e) shows the temperature evolutions of the mean-site fields. These were calculated taking the respective populations of the three muon sites into account. One immediately sees that the saturation mean field and the ordering temperature decrease with disorder.

For the $x = 0.15$ sample, the functional shape of the μ SR ZF spectra changes, and the three-site model is not applicable anymore. As seen in Fig. 6, the spectrum at this concentration

FIG. 6. (Color online) Zero-field μ SR time spectra of seven different concentrations. The data of $x = 0.3$ and $x = 0.5$ were taken at 0.02 K, and the data of $x = 0$, $x = 0.05$, $x = 0.15$, $x = 0.95$, and $x = 1$ were taken at 1.6 K. The solid lines are fit to the data described in the text. From bottom to top the spectra are offset by 0, 0.1, 0.2, 0.4, 0.5, 0.7, and 0.85.

FIG. 7. (Color online) (a)–(d) Measured temperature dependence of the internal fields at the three muon sites for (a) $x = 0$, (b) $x = 1$, (c) $x = 0.05$, and (d) $x = 0.95$ for the low-disorder samples (symbols) as described in the text. (e) Temperature dependence of the average internal magnetic field, assumed to be proportional to the order parameter (solid symbols). The open symbols are the average fields determined by Fourier transformation. In all cases, lines are guides for the eye.

shows a fast decay at early times, followed by a slow and weak oscillation. Denoting the oscillating fraction as A_1 and that with the fast decay as A_2 , for this concentration we used the following empirical fitting function:

$$A = \frac{2}{3}[A_1 \cos(\omega_1 t + \phi) \exp(-\lambda_1 t) + A_2 \exp(-(\lambda_2 t)^\beta)] + \frac{1}{3}(A_1 + A_2) \exp(-\lambda_{\text{tail}} t). \quad (3)$$

In these measurements, the background contribution was negligible. The ratio $A_1:A_2$ was fit globally with the resulting value of 1:1.336(4). In Eq. (3), ω_1 is the frequency of the oscillations. The phase ϕ was a global fit parameter for all temperatures yielding $\phi = 27(1)^\circ$. The parameter λ_1 is the damping rate of the oscillations. λ_{tail} is temperature independent and negligibly small as in the previously described concentrations. In Eq. (3) the second term represents all muons that stop in rather different magnetically environments. The stretched exponential is an attempt to account for the corresponding broad distribution of internal magnetic fields, which leads to a superposition of multiple harmonic precessions. The

FIG. 8. (Color online) Temperature dependence of the fitting parameters (a) λ and (b) β for highly disordered samples (symbols) as described in the text. Lines are guides for the eye.

temperature dependency of the parameters β and λ_2 is shown in Fig. 8. Upon lowering the temperature, a strong increase in λ_2 is observed at the transition point. Simultaneously, β changes in a steplike fashion from $\beta \sim 1.9$ to $\beta \sim 4.6$. The high-temperature value is in agreement with our expectation for the paramagnetic phase [22], whereas the high value at low temperature signals a broad unconventional distribution of local fields. The oscillation frequency ω_1 increases below the ordering temperature as expected.

For stronger disorder, in samples with $x = 0.3$ and $x = 0.5$, the oscillatory contribution becomes indiscernible. The corresponding data were analyzed using only the stretched exponential function,

$$A = \frac{2}{3}A_1 \exp[-(\lambda_1 t)^\beta] + \frac{1}{3}A_1 \exp(-\lambda_{\text{tail}} t) + A_{\text{bg}} \exp(-\lambda_{\text{bg}} t). \quad (4)$$

As for $x = 0.15$, the stretch exponential describes the superposition of a distribution of oscillating frequencies. The second term is again related to the muon spin component parallel to the internal field. The non-negligible background contribution is due to muons stopped in the ^3He -dilution refrigerator used in this run. A global fit for each sample gives $A_{\text{bg}} = 0.122(3)$ and $A_{\text{bg}} = 0.106(1)$ for $x = 0.5$ and $x = 0.3$. The temperature dependence of β and λ_1 is shown in Fig. 8. The exponent increases upon cooling through T_N . Its saturation values at the lowest temperature are rather similar: $\beta \approx 3.6$ for $x = 0.3$ and $\beta \approx 3.4$ for $x = 0.5$, respectively. In contrast, the relaxation rates λ_1 are about four times shorter for $x = 0.5$ as compared to $x = 0.3$.

3. Fourier inversion

Additional information was obtained using an alternative model-independent method to analyze the zero-field μ SR data. It is based on the fact that the muon time spectra are temporal Fourier transforms of the probability distribution of the muon spin Larmor precession frequencies in the sample [22]. Thus, a Fourier transform of the measured spectra will yield the probability distribution of the *modulus* of internal fields. Before applying this procedure to our data, we subtracted the tail and background contributions determined in model fits as described above. The result of the subsequent Fourier inversion is shown in Fig. 9. Both parent materials show two well-pronounced peaks. A third maximum can be identified as a shoulder of the higher-field peak. The central positions of these features correspond to the fitted field values in our model-based data analysis. At a 5% disorder on both ends we see a broadening of all features and a shift to smaller fields. At further higher concentrations the peaks merge, and the probability weight builds up at very small fields. For $x = 0.3$ and $x = 0.5$ the probability maxima are actually at zero field.

IV. DISCUSSION

It is clear from the above that chemical disorder in $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$ does not eliminate the well-defined magnetic ordering transitions at low temperatures. On the other hand, it has two obvious and rather drastic effects on the ordered state. First, disorder tends to suppress the average spontaneous static magnetization and the ordering temperature. Second, it results in a broad *distribution* of magnitudes of local fields (static moments) in the sample. We will address these two aspects of the observed behavior separately.

A. Average ordered moment

Despite the fact that several muon sites are involved, we will make the assumption that the average ordered moment is proportional to the average local field seen by the muons. Implicitly, we are assuming that the muon stopping sites are the same across the Ge-concentration range. In application to our data, the mean-local field is readily obtained from the measured probability distribution shown in Fig. 9. For $x = 0$, a moment of $m_0 = 0.108\mu_b$ can be estimated from chain-mean-field (MF) theory using the known values of J and T_N [11,23]. Note the strong suppression of m_0 by quantum fluctuations enhanced by the system's one dimensionality. Using the chain-MF estimate as a “calibration point” for the measured mean-local fields in other samples, in Fig. 4 we plot the saturation moment as a function of Ge concentration. Note the logarithmic y axis in this graph. As a consistency check, for the pure Ge sample, our procedure yields $m_0 = 0.072\mu_b$. This is only about 12% off the chain-MF estimate of $m_0 = 0.064$. One can thus hope that a 20% accuracy of our method holds for the entire Ge-concentration range.

Our analysis reveals a drastic suppression of the ordered moment already at very low levels of chemical randomness. This behavior emphasizes the quantum nature of the spin chains involved. In contrast, a classical antiferromagnet with fully saturated Néel sublattices would not be affected by bond

FIG. 9. (Color online) Measured internal magnetic-field distribution in $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$. The data of $x = 0.3$ and $x = 0.5$ were taken at $T = 0.02$ K. For $x = 0$, $x = 0.05$, $x = 0.15$, $x = 0.95$, and $x = 1$, $T = 1.6$ K.

disorder at all. Its ground state would remain the fully polarized two-sublattice state. In our case though, on the Si-rich end the decrease in m_0 is *exponential*. A similar (but somewhat less drastic) behavior was observed in the organic weakly coupled

FIG. 10. (Color online) Measured cumulative probability distribution of local fields in low-disorder $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$ samples. The x axis is normalized to the median value.

spin chain material $\text{Cu}(\text{py})_2\text{Cl}_{1-x}\text{Br}_x$ (CPX) [24]. At the same time, these trends totally contradict chain-MF calculations for bond-disordered chains where both T_N and m_0 were predicted to increase [25].

B. Distribution of ordered moment

The most important result of this paper is the study of the *distribution* of ordered moments in samples with bond randomness. This kind of analysis was not possible for our previous experiments on CPX [24]. In the case of $\text{BaCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2\text{O}_7$, already the fact that the data are described by a stretched exponential relaxation with a large exponent β indicates a very unconventional distribution of local fields. In fact, a substantial broadening of the field distribution is already present at very small disorder levels. The most unbiased way to visualize this is by integrating the measured probability distributions as shown in Fig. 10. Here the scale of the x axis for each material is chosen so as to have their median field points coincide. We see that the magnitudes of local fields in both weakly disordered samples with $x = 0.05$ and $x = 0.95$ have a much broader relative distribution than in the parent material. In particular, the proportion of relatively large fields is substantially increased by disorder.

As mentioned above, the ordered moment in weakly coupled spin chains is suppressed by quantum spin fluctuations. Any bond strength disorder will modulate the strength of these quantum fluctuations in a spatially random manner. The result will be an inhomogeneous spatial distribution

of ordered moments. However, it is clear that at least in our low-disorder samples, the inhomogeneity is not confined to the rare and isolated substitution sites. Indeed, for $x = 0.05$ and $x = 0.95$, the vast majority of muons rest in a relatively homogeneous environment, similar to that in the corresponding parent compound. Rather, the broad distribution as well as a disproportionate prevalence of relatively large local fields (moments) are due to the RS nature of the individual chains.

Consider a single random-bond chain. In the ordered phase, at the MF level, it is subject to a staggered field generated by neighboring chains. The RS ground state can be viewed as a collection of *noninteracting* effective spin dimers [2–4]. The probability distribution of the corresponding dimer coupling strengths J is peaked at zero, i.e., there is an abundance of very weak effective dimers that also will tend to involve physically distant spins [2–4]. Each such dimer will get polarized by the staggered mean field. The resulting induced moment will be proportional to the dimer’s susceptibility, which, at low temperatures, is inversely proportional to J . Thus, the distribution of magnetic moments will echo the distribution of inverse exchange constants. It will be very broad and will have a disproportionate number of large moments. These are due to the particularly weak dimers in the RS state. Unfortunately, to date, neither the probability distribution nor the spatial distribution of ordered moments in weakly coupled RS chains have been addressed theoretically or numerically.

The inhomogeneous ground state will undoubtedly affect the low-energy excitations. A hint of this phenomenon is present in our calorimetry data. In the presence of disorder, quasimomentum ceases to be a good quantum number, and the entire spin wave excitation picture breaks down. The observed decrease in the effective $C \propto T^\alpha$ power-law exponent signals a disproportional abundance of low-energy excitations. A decrease in specific heat is directly related to a modification. At this point we can only speculate that this is a consequence of the RS nature of the individual chains too. Undoubtedly more insight will come from further theoretical studies.

V. CONCLUSION

To summarize, even weak bond strength disorder strongly affects magnetic ordering in weakly coupled quantum spin chains. The average ordered moment is drastically suppressed. At the same time the distribution of local static moments is highly inhomogeneous with a disproportionate number of relatively large moments. The thermodynamics of the ordered state is also affected. All this appears to be a direct consequence of “infinite disorder” in individual random-bond chains.

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