Antiferromagnetic order in weakly coupled random spin chains

J. Kokalj 1, J. Herbrych 2, A. Zheludev 3, and P. Prelovšek 1,4

1 J. Stefan Institute, SI-1000 Ljubljana, Slovenia
2 Crete Center for Quantum Complexity and Nanotechnology, Department of Physics, University of Crete, P.O. Box 2208, 71003 Heraklion, Greece
3 Neutron Scattering and Magnetism, Laboratory for Solid State Physics, ETH Zürich, Zürich, Switzerland
4 Faculty of Mathematics and Physics, University of Ljubljana, SI-1000 Ljubljana, Slovenia

(Dated: September 10, 2014)

The ordering of weakly coupled random antiferromagnetic \( S = 1/2 \) chains, as relevant for recent experimentally investigated spin chain materials, is considered theoretically. The one-dimensional isotropic Heisenberg model with random exchange interactions is treated numerically on finite chains with the density-matrix renormalization-group approach as well as with the standard renormalization analysis, both within the mean-field approximation for interchain coupling \( J_\perp \). Results for the ordering temperature \( T_N \) and for the ordered moment \( m_0 \) are presented and are both reduced with the increasing disorder agreeing with experimental observations. The most pronounced effect of the random singlet concept appears to be a very large span of local ordering moments, becoming wider with decreasing \( J_\perp \), consistent with μSR experimental findings.

PACS numbers: 05.60.Gg, 25.40.Fq, 71.27.+a, 75.10.Pq

The antiferromagnetic (AFM) Heisenberg model of \( S = 1/2 \) spins on a one-dimensional (1D) chain represents one of the prototype and most studied quantum many-body model for strongly correlated electrons, being at the same time realized nearly perfectly in several materials. Since 1D spin systems do not exhibit any long range order even at temperature \( T = 0 \), the ordering can appear through the interchain coupling. The ordering Néel temperature \( T_N \) emerging in weakly coupled AFM chains is by now well described theoretically \([1]\), being confirmed by numerical calculations \([2]\) and the experimental investigations on materials with quasi-1D spin systems \([3]\).

The quenched disorder in intrachain exchange couplings \( J_i \) reveals in 1D spin chains qualitatively new phenomena as well theoretical and experimental challenges. Even in the case of unfrustrated AFM random Heisenberg chain (RHC) with \( J_i > 0 \) it has been shown using the renormalization-group (RG) approaches \([4-7]\) that the \( T \to 0 \) behavior is qualitatively changed by any disorder leading to the concept of random singlets (RS). The signature of such state is the singular - Curie-like - divergence of the uniform susceptibility \( \chi_0(T \to 0) \) \([8]\) being an indication of the vanishing effective exchange coupling \( J_{\text{eff}} \). Refreshed theoretical interest in RHC phenomena has been stimulated by the synthesis and experimental investigations of novel materials representing the realisation of RHC, in particular BaCu\(_2\)(Si\(_{1-x}\)Ge\(_x\))\(_2\)O\(_7\) \([3, 9, 10]\) and Cu(py)\(_2\)(Cl\(_{1-x}\)Br\(_x\))\(_2\) \([11]\) compounds. Experiments confirmed theoretically predicted \( \chi_0(T) \) \([12]\), but revealed also novel features as large and strongly \( T \)-dependent spread of local NMR spin-lattice relaxation times \([10, 13]\) which has been reproduced within the simple model of 1D RHC \([14]\).

The existence of weak but finite interchain couplings \( J_\perp \) in quasi-1D RHC compounds and related AFM ordering at low \( T < T_N \) open a new perspective on the RS systems \([11]\). Mixed BaCu\(_2\)(Si\(_{1-x}\)Ge\(_x\))O\(_7\) \([9]\) as well Cu(py)\(_2\)(Cl\(_{1-x}\)Br\(_x\))\(_2\) \([11]\) show a substantial reduction of \( T_N \) as well as the ground state (g.s.) \( T = 0 \) ordered magnetic moment \( m_0 \) relative to the disorder-free \( x = 0 \) and \( x = 1 \) materials. Theoretical treatments so far suggested even the opposite trend \([15]\) revealing the difficulties of theoretical approaches. The central theoretical issue also in connection with experiments is to what extent and in which properties the singular behavior of quantum RS physics remains reflected in the long-range AFM order at low \( T \). The aim of this Letter is to present results of numerical and analytical calculations which show that under the presence of weak (but not extremely weak) interchain coupling treated within a mean-field approximation (MFA) randomness reduces both \( T_N \) as well as \( m_0 \), which is in agreement with experiment. We also present evidence that the RS phenomena are reflected in large distribution of \( T = 0 \) local ordered moments \( m_i \) being consistent with preliminary experimental results \([16]\).

Our goal is to understand properties in particular the ordering in the quasi-1D RHC model, which is given by quenched (intrachain) random exchange couplings \( J_{i,j} \) and constant interchain coupling \( J_\perp \),

\[
H = \sum_{i,j} J_{i,j} \mathbf{S}_{i,j} \cdot \mathbf{S}_{i+1,j} + J_\perp \sum_{i,j'\neq j} \mathbf{S}_{i,j} \cdot \mathbf{S}_{i,j'},
\]

where \( \mathbf{S} = S = 1/2 \) spin operators. The isotropic Heisenberg coupling is assumed both within the chain \( (J_{i,j} \) with \( i \) denoting sites in the chain and \( j \) denoting different chains) as well as for the interchain term \( (j,j') \) run over \( z_\perp \) nearest-neighbor chains. Since we are discussing possible ordering at low \( T \) the only reasonable starting point is the MFA for the interchain coupling. In actual compounds the spin system is close to two-dimensional with modest \( z_\perp = 2 \) as well with less clear role of disorder on \( J_\perp \) which we discuss again in conclusions. Still we expect in analogy to other quasi-1D spin systems \([1, 2]\) that main ordering features should be captured by the effective 1D RHC with the staggered field \( h_s \) provided...
that $J_\perp \ll J_i$,

$$H_{\text{MF}} = \sum_i J_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} - h_s \sum_i (-1)^i S^z_i. \tag{2}$$

Within the MFA the staggered field is given by $h_s = z_\perp J_\perp m_s$ with the staggered magnetization $m_s = (1/L) \sum | \langle \ldots \rangle |$ and $\langle \ldots \rangle$ denoting thermal average. We will further on consider random quenched $J_i$ and for convenience assume their distribution to be uncorrelated uniform boxed distribution with $J - \delta J \leq J_i \leq J + \delta J$ and $\delta J < J$. For experimental examples more appropriate distribution would be binary one, but it has been verified [14] that qualitative features do not depend essentially on the form of the distribution.

In the following presentation of results we use units $J = 1$ and set $k_B = \hbar = 1$.

Within the MFA for the interchain coupling the instability towards the AFM ordering and the ordering temperature $T_N$ are determined by the staggered static susceptibility $\chi_\pi$ of a 1D chain and the relation [1, 17]

$$z_\perp | J_\perp | \chi_\pi(T_N) = 1. \tag{3}$$

We evaluate $m_s(T)$ and $\chi_\pi(T)$ using the finite-temperature dynamical matrix renormalization group (FTDMRG) method [18, 19] on a finite chain with $L$ sites and open boundary conditions. In this method standard $T = 0$ DMRG targeting of ground state density matrix $\rho^0 = |0\rangle \langle 0|$ is generalized with finite-$T$ density matrix $\rho^T = (1/Z) \sum_r \langle \ldots | e^{-H/T} | \ldots \rangle$. Next, the reduced density matrix is calculated and then truncated in the standard DMRG-like manner for basis optimization. The limitation of the FTDMRG method are at low $T$ finite-size effects, which are rather small due to large accessible system with DMRG algorithm and which are even further reduced with randomness. For systems with $\delta J > 0$ we employ also random configuration averaging, typically over $N_r = 10$ realizations for finite- $T$. For $T = 0$ we use smaller $N_r = 5$, since standard DMRG method and larger systems can be used. $\chi_\pi$ can be evaluated via dynamical susceptibility $\chi^{\rho}(\pi, \omega)$, still we use mostly the alternative approach by evaluating $m_s$ at finite $T$ and $h_s$, and then using $\chi_\pi(T) = \lim_{h_s \to 0} m_s(T, h_s)/h_s$. Within this approach numerical results are more robust or reliable since only static quantities are calculated and finite size or boundary effects can be reduced, e.g., by considering only sites close to the middle of a chain. Still, limit $h_s \to 0$ is hard to reach numerically, but at finite $T$ small field $h_s \sim 0.01$ suffices [19].

Results for $\chi_\pi$ used to extract $T_N$ with Eq. (3) are for several $\delta J$ shown in Fig. 1a. For $\delta J = 0$ analytical approaches [20–22] suggest that $T \to 0$

$$\chi_\pi^{\rho} = a \sqrt{\log(b/T)}/T, \tag{4}$$

for which quantum Monte Carlo approach gives [23, 24] $a = 0.32, 0.30$ and $b = 5.9, 9.8$. Results for random $\delta J \neq 0$ shown in Fig. 1a clearly indicate that the increasing $\delta J$ reduces $\chi_\pi$ and consequently leads to a systematic decrease of

$$T_N = 0.0001 \leq \delta J \leq 0.8.$$
son to the analytical result [1],
\[ m_s^0 = r(h_s)^r. \]  \( \text{(6)} \)

This result with \( r = 0.637 \) and \( g = 1/3 \) is obtained by elimination of \( J_2 \) from Eq. (7) and MFA connection \( h_s = -4J_1m_0 \) used in Ref. [1]. In Fig. 2b we compare Eq. (6) to our DMRG results and reveal substantial differences. Our \( m_s(h_s) \) for \( dJ = 0 \) shows rather stronger increase with \( h_s \) (and therefore larger \( m_s \) at low \( h_s \)), which cannot be reconciled with Eq. (6) simply by just an increase of prefactor \( r \).

Linear dependence of \( m_s(h_s) \) for \( dJ = 0 \) in Fig. 2b suggests different exponent (\( g \neq 1/3 \)) or possibly some logarithmic corrections.

Results in Fig. 2a,b show that disorder \( \delta J \) leads to a decrease of staggered magnetization \( m_s \) in our \( h_s \)-regime. A possibility of increased \( m_s \) with increased \( \delta J \) remains at very low \( h_s < 0.0001 \). This is suggested by results in Fig. 2b and we investigate and discuss it later also with the use of RG method. The \( T = 0 \) solutions of the MFA self-consistency relation \( h_s/(z_\perp J_\perp) = m_s(h_s) \) give ordered moment \( m_0 \) and we present its decrease with \( \delta J \) for different values of \( z_\perp J_\perp \) in Fig. 2c.

A novel feature introduced by disorder is the distribution of local ordered moments. To avoid the influence of open boundary conditions we calculate local staggered \( m_i = (-1)^i\langle S_i^z \rangle \) from the middle of the chain modeled with Eq. (2) and for the MFA self-consistent fields \( h_s \) at particular \( z_\perp J_\perp \). Even in a uniform staggered field \( h_s \) moments \( m_i \) are found to vary from site to site and depend on the concrete random configuration \( J_i \). We present the probability distribution function (PDF) in Fig. 3a for different randomness \( \delta J \) and fixed \( z_\perp J_\perp = 0.05 \), while in Fig. 3b we show it for fixed \( \delta J \) and different \( z_\perp J_\perp \). It is evident from Fig. 3a that for large disorder and small \( z_\perp J_\perp \) the PDF largely deviate from the Gaussian-like form. Moreover, the relative spread of distribution \( \Delta = \sigma(m_i)/m_0 \) can become even \( \Delta > 1 \).

For better understanding and interpretation of above results within the RS concept we perform similar real space renormalization group procedure to the one introduced by Dasgupta and Ma [5], in which bonds with largest exchange couplings are eliminated and reduced effective coupling \( J_{\text{eff}} \) introduced. We generalized the procedure for finite \( h_s \) and for calculation of \( m_0 \) and give more technical details of the procedure, which is similar to the one used in Ref. [15], in the Supplemental material [19]. We perform RG procedure numerically on a large system and by carrying it to the end together with evaluation of staggered magnetization for different starting staggered fields we obtain \( m_s(h_s) \) for \( T = 0 \). A simple RS argument suggest that in a finite \( h_s \) all spins with effective coupling \( J_{\text{eff}} < h_s \) are fully polarized, while the ones with \( J_{\text{eff}} > h_s \) form singlets and contribute only weakly to the staggered magnetization. Since the portion of spins with \( J_{\text{eff}} < h_s \) in a RS theory is \( \propto \ln^{-2}(n/h_s) \) [5], one expects for small \( h_s \)

\[ m_s^{RS}(h_s) \propto \ln^{-2}(n/h_s). \]  \( \text{(7)} \)

Figure 2. (Color online) (a) \( T = 0 \) staggered magnetization \( m_s \) vs. \( h_s \) for various randomness \( \delta J \), calculated for \( L = 800 \) and \( N_e = 5 \). (b) Log-log plot of \( m_s \) vs. \( h_s \) for \( \delta J = 0, 0.8 \). \( m_s(h_s) \) for \( \delta J = 0 \) deviates from prediction in Eq. (6) in exponent \( d \) and prefactor \( c \). The result for \( \delta J = 0.8 \) shows a RS like behaviour given with Eq. (7). Fits of parameters for Eq. (6) or (7) are for regime \( 0.0001 < h < 0.01 \). (c) Self-consistent solution for staggered magnetization \( m_0 \) vs. \( \delta J \) for different \( z_\perp J_\perp \).

We confirm this RS prediction with numerical RG results [19]. Our \( T = 0 \) DMRG results also confirm such behavior as shown in Fig. 2b at low \( h_s \), since they deviate from simple power law behaviour of Eq. (6) (linear in log-log plot) with a substantial upward curvature, nicely captured with Eq. (7). Therefore our result in Fig. 2b represents one of a few [13, 25, 26] confirmations of the RS phenomenology.

From RG procedure one can make also predictions for finite-\( T \) results, which are usually [5, 6] obtained by preforming RG steps as long as some Hamiltonian parameter (e.g. exchange coupling) is larger than \( T \), while for the remaining system with all effective parameters below \( T \), a high \( T \) result is used. We apply similar procedure for \( m_s(h_s) \) at finite \( T \) (see Supplement for details [19]) and obtain the RS prediction for the susceptibility in Eq. (5). This has the same
Figure 3. (Color online) Probability distribution function of $m_i$ for (a) various values of $\delta J$ and fixed $z_\perp J_\perp = 0.05$, and (b) for fixed $\delta J = 0.4$ and various $z_\perp J_\perp$, as calculated for $L = 800$, $T = 0$ and $N_r = 5$. Thin, vertical lines represent $m_0$ for given $\delta J$ and $J_\perp$.

A functional form as a RS prediction for uniform susceptibility $[6,8] \chi_0(T)$, which can be expected for random system with no translational symmetry and strongly local correlations. In Fig. 1a we show that our numerical calculations with FTD-DMRG give support to this RS prediction.

Summarizing our theoretical and numerical results, we conclude that at fixed average $\langle J \rangle$ and interchain coupling $J_\perp$, the disorder $\delta J > 0$ leads to a decrease of Néel temperature $T_N$ as well as to reduced g.s. ordered staggered moment $m_0$, in a very broad range of $\delta J > 0$ (at least in the range evaluated in our study). This is due to $\chi_\pi$ being smaller for random system than for a pure system in a relevant regime (see Fig. 1a), which is in contrast with the uniform case $q = 0$ susceptibility $\chi_0(T \rightarrow 0)$ which approaches constant for pure case but diverges $\propto 1/[T \ln^2(\beta/T)]$ for $\delta J > 0$. This is analogous to Eq. (5) and a direct signature of RS scenario leading at low $T \rightarrow 0$ to formation of local singlets and almost free spins. The effect of disorder at $q = \pi$ is less dramatic since to the leading order (neglecting log corrections) both ordered and $\delta J > 0$ cases reveal $\chi_\pi \propto 1/T$.

Numerical results for $m_\perp(h_\perp)$ at $T = 0$ in Fig. 2(a,b) show that in the regime with larger $h_\perp$ (e.g., $h_\perp > 0.0001$ for $\delta J = 0.8$) the average moment $m_\perp$ (and in turn $m_0$ shown in Fig. 2c) decreases with increasing $\delta J$. On the other hand, Eqs. (6), (7) and results in Fig. 2 suggest a regime of very low $h_\perp$ where $m_\perp (m_0)$ could be increased by $\delta J > 0$. This could be only relevant for larger $\delta J$ and for very small $J_\perp (\ll 0.001$ for $\delta J = 0.8$) which would lead to enhanced $T_N$ and $m_0$ with increased $\delta J$ or in other words, to order by disorder. Such behavior was actually predicted by MFA and RG treatment [15], but is contrary to the one mainly discussed here, as well not found in materials of interest [16].

The most striking effect of the RHC physics and of anomalous RS response in the ordered phase is however the distribution of local moments $m_\perp$, as manifested by PDF($m_\perp$) in Fig. 3. It is evident that the relative distribution width $\Delta$ of the distribution $m_\perp/m_0$ increases with $\delta J$ but even more important with decreasing $z_\perp J_\perp$. It should be noted that for larger $\delta J$ even $m_\perp < 0$ becomes possible (moments $m_\perp$ locally opposite to local fields) [13]. This means that at small $J_\perp \ll J$ and strongly reduced $T_N$ the PDF width can become large, i.e. $\Delta \sim 1$. We note also that within our MFA analysis the constant average staggered field $h_\perp$ was used. Taking into account also the local fluctuation of $h_\perp$ will necessarily lead to the even further increase of $\Delta$, and possibly to even further reduction of $T_N$ and $m_0$. For a pure system it has also been shown [2] that going beyond MFA results in an effectively reduced $z_\perp$, which would also lead to reduced estimates for $T_N$ and $m_0$.

Turning to the experimental realizations of random spin chains, two systems have been studied so far with magnetic ordering at low $T$, namely BaCu$_2$(Si$_{1-x}$Ge$_x$)$_2$O$_7$ [9, 10, 16] and Cu(py)$_2$(Cl$_{1-x}$Br$_x$)$_2$ [11]. The former material is more relevant to the present discussion, since a clear evidence of 1D RS physics has been detected there for $T > T_N$, i.e. above the 3D ordering [10]. Its magnetic properties can be well described by a simple bimodal distribution of in-chain exchange constants [10]. In this model, the spin chains have two intrachain random quenched AFM $J_1 = J_2$, with probabilities $x$ and $1 - x$, respectively, and weak interchain coupling $J_\perp < J$. Although our treatment assumes a uniform distribution of the exchange constants, it should be able to capture general features of BaCu$_2$(Si$_{1-x}$Ge$_x$)$_2$O$_7$, particularly with Ge concentration $x \sim 0.5$ [14].

The experimental data that are most relevant to our calculations are $\mu$-SR experiments, from which the magnitude of $m_0$ can be inferred. In full agreement with our predictions, in both Cu(py)$_2$(Cl$_{1-x}$Br$_x$)$_2$ [11] and BaCu$_2$(Si$_{1-x}$Ge$_x$)$_2$O$_7$ [16], $m_0$ and the ordering temperature $T_N$ were found to decrease with increasing disorder. This said, the drop in BaCu$_2$(Si$_{1-x}$Ge$_x$)$_2$O$_7$ appears much more abrupt than predicted. This may be an indication of limitations for the chain-MF approach, but may also be related to the observation [9], that the inter-chain coupling strength and even its sign may be locally affected by disorder.

The most interesting experimental observation for BaCu$_2$(Si$_{1-x}$Ge$_x$)$_2$O$_7$ is a drastic broadening probability distribution of the local static moments in the magnetically ordered state [16]. This behavior is totally consistent with our predictions borne in Fig. 3. Unfortunately, making a quantitative comparison beyond a qualitative agreement is not feasible at the present stage. The problem is that $\mu$-SR actually measures the distribution of local magnetic fields, not magnetic moments. Due to the presence of several crystallographic muon sites, the $m_0$ distribution can not be unambiguously extracted from such experiments.

We acknowledge helpful and inspiring discussions with M. Thede. We acknowledge the support of the European Union.
program (J.H.) FP7-REGPOT-2012-2013-1 no. 316165 and of the Slovenian Research Agency under program (P.P.) P1-0044 and under grant (J.K.) Z1-5442.

[1] H. J. Schulz, Phys. Rev. Lett. 77, 2790 (1996).
[2] C. Yasuda, S. Todo, K. Hukushima, F. Alet, M. Keller, M. Troyer, and H. Takayama, Phys. Rev. Lett. 94, 217201 (2005).
[3] I. Tsukada, Y. Sasago, K. Uchinokura, A. Zheludev, S. Maslov, G. Shirane, K. Kakurai, and E. Ressouche, Phys. Rev. B 60, 6601 (1999).
[4] S.-k. Ma, C. Dasgupta, and C.-k. Hu, Phys. Rev. Lett. 43, 1434 (1979).
[5] C. Dasgupta and S.-k. Ma, Phys. Rev. B 22, 1305 (1980).
[6] D. Fisher, Phys. Rev. B 50, 3799 (1994).
[7] E. Westerberg, A. Furusaki, M. Sigrist, and P. A. Lee, Phys. Rev. Lett. 75, 4302 (1995).
[8] J. Hirsch, Phys. Rev. B 22, 5355 (1980).
[9] T. Yamada, Z. Hiroi, and M. Takano, J. Solid State Chem. 156, 101 (2001).
[10] T. Shiroka, F. Casola, V. Glazkov, A. Zheludev, K. Prša, H.-R. Ott, and J. Mesot, Phys. Rev. Lett. 106, 137202 (2011).
[11] M. Thede, F. Xiao, C. Baines, C. Landee, E. Morenzoni, and A. Zheludev, Phys. Rev. B 86, 180407 (2012).
[12] A. Zheludev, T. Masuda, G. Dhalenne, A. Revcolevschi, C. Frost, and T. Perring, Phys. Rev. B 75, 054409 (2007).
[13] T. Shiroka, F. Casola, W. Lorenz, K. Prša, A. Zheludev, H.-R. Ott, and J. Mesot, Phys. Rev. B 88, 054422 (2013).
[14] J. Herbjörnsson, J. Kokalj, and P. Prelovšek, Phys. Rev. Lett. 111, 147203 (2013).
[15] A. Joshi and K. Yang, Phys. Rev. B 67, 174403 (2003).
[16] M. Thede, T. Haku, T. Masuda, C. Baines, E. Pomjakushina, G. Dhalenne, A. Revcolevschi, E. Morenzoni, and A. Zheludev, ArXiv e-prints (2014), arXiv:1407.0813 [cond-mat.str-el].
[17] D. J. Scalapino, Y. Imry, and P. Pincus, Phys. Rev. B 11, 2042 (1975).
[18] J. Kokalj and P. Prelovšek, Phys. Rev. B 80, 205117 (2009).
[19] See Supplemental Material for more details.
[20] T. Giamarchi and H. Schulz, Phys. Rev. B 39, 4620 (1989).
[21] R. R. P. Singh, M. E. Fisher, and R. Shankar, Phys. Rev. B 39, 2562 (1989).
[22] I. Affleck, D. Gepner, T. Ziman, and H. J. Schulz, J. Phys. A: Math. Gen. 22, 511 (1989).
[23] O. A. Starykh, A. W. Sandvik, and R. R. P. Singh, Phys. Rev. B 55, 14953 (1997).
[24] Y. Kim, M. Greven, U.-J. Wiese, and R. Birgeneau, Eur. Phys. J. B 4, 291 (1998).
[25] J. Hoyos, A. Vieira, N. Laflorencie, and E. Miranda, Phys. Rev. B 76, 174425 (2007).
[26] K. Hida, J. Phys. Soc. Jpn. 65, 895 (1996).
I. REAL SPACE RENormalization Group

I.a. Procedure

We numerically performed similar renormalization group procedure as introduced by Dasgupta and Ma [S1] and modified it to include the staggered magnetic field $h_s$ and extended it for calculation of staggered magnetization $m_s$, similarly as done in Ref. [S2]. In the original procedure the bonds with largest $J_i$ were eliminated which we replace by subsequent elimination of bonds with largest $J_{xx,i}$. In the presence of broken rotational symmetry due to staggered magnetic field $h_s$, $J_{xx,i}$ does not equal $J_{zz,i}$ at further steps of the elimination process. In the case of $h_s = 0$ the criteria equals to the original one used by Dasgupta and Ma [S1] and $J_{xx,i} = J_{zz,i}$. Justification of $J_{xx,i}$ for elimination criteria is also that it is the only non-diagonal element of the Hamiltonian and that for $J_{xx,i} = 0$ the ground state is a simple product state or Neel state, which can be exactly obtained by arbitrary order of the elimination steps provided that elimination is performed to the end. For finite-$T$ properties also other energy scales like $J_{xx,i}$ and $h_s$ are important and need to be considered.

Once the bond of two sites to eliminate are chosen we integrate them out by the following procedure. First we calculate eigenstates of the four site Hamiltonian which consists of two sites to be eliminated (namely sites 2 and 3) plus two neighboring sites (namely sites 1 and 4). Usually the relevant states which we would like to keep are the four lowest states and from which we could build effective Hamiltonian or the new bond (from site 1 to 4) parameters. However, as the elimination procedure advances the four lowest states of the four site Hamiltonian do not necessarily have the character of the ground state on eliminated bond (sites 2, 3) i.e. they do not all have large overlap with it and some state with the character of higher lying state on sites 2 and 3 might become low and among first four low lying states of the four site Hamiltonian.

This does not happen if $J_{12}^{zz,zz}$ and $J_{14}^{zz,zz}$ are much smaller than $J_{23}^{zz,zz}$. In such case we choose four eigenstates of the 4 site Hamiltonian with the largest overlap with the ground state on eliminated two sites (sites 2, 3). These four states span the part of the relevant low energy Hilbert space that we would like to keep and are close to the states kept in the second order procedure in Ref. [S1].

From this four states ($\{\psi_i\}$ with energy $E_i$, $i = 1, \ldots, 4$) we build new effective Hamiltonian for the remaining sites (sites 1, 4) by first constructing $H_{1234} = \sum_i |\psi_i\rangle E_i \langle\psi_i|$ and then tracing out the eliminated sites $H_{14} = \sum_{i|23}|\psi_{23}\rangle H_{1234}|\psi_{23}\rangle$. Here $|\psi_{23}\rangle$ are basis states for eliminated sites (sites 2, 3). New $H_{14}$ is the new Hamiltonian in the basis of remaining sites (1 and 4) and from which one can read new effective parameters like $J_{14}^{xx}, J_{14}^{zz}, h_1, h_4$ and energy of integrated out sites $E_{23}$.

Similar procedure can be used for determining the parameters of new operators that we are interested in. For example, operator $a_1 S_{11}^z + a_2 S_{22}^z + a_3 S_{33}^z + a_4 S_{44}^z$ is transformed into new operator $\tilde{a}_1 S_{11}^z + \tilde{a}_4 S_{44}^z + o_{23}$ after integrating out sites (2 and 3), while in this case the parameters $a_1, \tilde{a}_4$ and $o_{23}$ need to be optimally chosen and small relative error (typically of $10^{-6}$) can appear by approximating the operator in the basis for remaining sites (1 and 4) by just three parameters.

In this way one eliminates the two sites, obtains new effective parameters for the Hamiltonian and operator on the new bond (connecting site 1 and 4) and can proceed with the new step of RG or by choosing next two sites to eliminate.

The ground state energy and expectation value of the operator in the ground state are obtained by preforming the RG to the end (eliminate all sites) and summing all $E_{23}$ and $o_{23}$ for the energy and the operator expectation values, respectively.

In Fig. S1 we show that our numerical RG decimation reproduces the random-singlet result of exponential decrease of the maximal $J$ in the system, $J_{\max}(L) = a \exp(bL/L_\text{r})$, as the RG procedure proceeds. $L_\text{r}$ is renormalized length, $L$ is full length of the system and $J_{\max}$ is the maximal $J$ in the renormalized system of length $L_\text{r}$. This behaviour is responsible for logarithmic corrections to the divergence of the uniform spin susceptibility at low $T$, $\chi_0(T) \propto 1/[T \ln^3(b/T)]$ [S3].

In Fig. S2 we show variation of parameters with RG steps in the presence of staggered magnetic field $h_s$.

I.b. $T = 0$ results

Carrying RG to the end and evaluating contribution of all eliminated sites to the staggered magnetisation (as demonstrated with $\tilde{m}_s$ in Fig. S2) we get the total magnetization $m_s(T = 0)$, which equals $\tilde{m}_s(L_\text{r} = 0)$. By performing RG for different $h_s$ we obtain $h_s$ dependence of $m_s(T = 0)$, which follows the simple RS argument and is shown in Fig. S3.
Our RG reproduces the random singlet result of exponentially decreasing coupling constant with increasing number of RG steps or decreasing renormalized length of the system \( L_r \). Random singlet behaviour appears only for very small values of coupling constant (< 0.01) and at small relative renormalized system sizes \( L_r/L < 0.1 \) since many RG steps are needed to reach asymptotic behaviour. Jumps in \( J_{\text{max}}(L_r) \) are remnants of the edges of initial box \( J \) distribution. Fit is the random singlet prediction \( J_{\text{max}}(L_r) = a \exp(b L_r/L) \) with adjusted \( a \) and \( b \). Results are for \( L = 100000, \delta J = 0.8 \) and no magnetic field.

\[ J_{\text{max}} \]

\[ L_r/L \]

\[ J_{\text{max}} \]

\[ \text{RS fit} \]

Figure S1. Our RG reproduces the random singlet result of exponentially decreasing coupling constant with increasing number of RG steps or decreasing renormalized length of the system \( L_r \). Random singlet behaviour appears only for very small values of coupling constant (< 0.01) and at small relative renormalized system sizes \( L_r/L < 0.1 \) since many RG steps are needed to reach asymptotic behaviour. Jumps in \( J_{\text{max}}(L_r) \) are remnants of the edges of initial box \( J \) distribution. Fit is the random singlet prediction \( J_{\text{max}}(L_r) = a \exp(b L_r/L) \) with adjusted \( a \) and \( b \). Results are for \( L = 100000, \delta J = 0.8 \) and no magnetic field.

**Lc. Finite \( T \) results**

To obtain finite \( T \) results within RG one usually performs RG steps as long as integrated out sites have energy scale larger than \( T \), while for the rest of the system high \( T \) result is used. In our case with the system in finite magnetic field \( h_s \), this fields do not get reduced with RG and therefore roughly set the lowest energy scale (see Fig. S2). This means that for \( T < h_s \) one can perform the RG to the end and obtain \( T = 0 \) result for all \( T < h_s \). Once \( T \) becomes above \( h_s \), all steps with \( J < h_s \) cannot be performed (for \( L_r/L < 0.12 \) in Fig. S2) and for this remaining system the high-\( T \) result (\( m_s \) roughly linear in \( h_s \)) should be used. This would lead to random singlet like prediction of for \( h_s \ll T \),

\[ m_s = h_s \frac{a}{T \ln^2(b/T)} \]  

(S1)

with \( a \) and \( b \) comparable to the ones for the RS fit in the inset of Fig. S3. RG therefore predicts similar functional form for the staggered susceptibility as for the uniform susceptibility \( \chi_0 \).

**II. DENSITY MATRIX RENORMALIZATION GROUP**

**II.a. Procedure**

Here we give just a short overview of the algorithm, since the detailed description of the numerical method can be found in Ref. S5 and also in Supplementary material of Ref. S6.

The quenched random \( J_i \) are introduced into the DMRG procedure at the beginning of finite algorithm. *Infinite algorithm is preformed for homogeneous system \( J_i = J \) and the randomness of \( J_i \) is introduced in the first sweep. In this way the preparation of the basis in the infinite algorithm is performed just once and for all realizations of \( J_i \)-s, while larger number of sweeps (usually \( \sim 5 \)) is needed to converge the basis within the finite algorithm for random \( J_i \). After finite algorithm magnetization \( \langle S_i^z \rangle \) at desired \( T \) is calculated at every site of the chain within measurements part of DMRG procedure.

**II.b. \( T = 0 \) results**

Since we are interested in a static quantity, namely ground state magnetization \( m_s(h_s, T = 0) \), large system sizes \( L > 100 \) can be used for \( T = 0 \) DMRG method. In Fig. S4 we present the system size dependence of \( m_s \) for pure system \( \delta J = 0 \), which has largest finite size effects, and show that the convergence with \( L \) can be reached to quite low \( h_s \) (of the order of \( 10^{-4} \)).
Finite temperature calculations are more demanding. The FTD-DMRG method is most efficient at low-\( T \), where the basis is more efficiently truncated and only small portion (\( M \) basis states) of the whole basis per block can be kept with small truncation error. We typically keep \( M \sim 200 \) basis states in the DMRG block and use systems with length \( L \sim 80 \).

In Fig. S5 we present the finite size dependence of \( m_s/h_s \) for low fields (\( h_s = 0.01 \)) used for evaluation of \( \chi_\pi \) to show good system size convergence in the presented regime of finite \( T \). We show results for most demanding pure system (\( \delta J = 0 \)) and one random system with \( \delta J = 0.6 \).

In Fig. S6 we show the dependence of \( m_s/h_s \) on \( h_s \) at finite \( T \), which saturates at low \( h_s \) and saturated value corresponds to \( \chi_\pi \). For pure case with \( \delta J = 0 \) such saturation with decreasing \( h_s \) can be reached for \( T \geq 0.1 \) as shown in Fig. S6(a,c) while for random case with \( \delta J = 0.6 \) it can be reached even at lower \( T \) (\( \cong 0.05 \)) (see Fig. S6(b,d)).

For an overall behaviour and for completeness we show in Fig. S7 \( m_s(h_s) \) for several \( T \) in a broader \( h_s \) regime.

**Figure S3.** (Color online) Staggered magnetization \( m_s \) at \( T = 0 \) vs. staggered magnetic field \( h_s \) as obtained with RG. Magnetization for random system (\( \delta J = 0.8 \)) is compared with the Schulz’s result [S4]. Eq. (5) in main text, for pure Heisenberg chain. \( m_s \) in a random system is typically smaller than for pure system except at very low fields (\( h_s < 0.01 \)). This can be understood with extension of random singlet result implying that \( m_s = a \ln^{-2}(b/h_s) \) and describes RG results better than power law (\( \propto h_s^\alpha \)) behaviour (see inset). The strong increase of \( m_s \) at low \( h_s \) (\(< 0.01 \)) originates in asymptotically free spins or strongly reduces effective coupling \( J_{\text{eff}} \) at the final stages of the RG procedure [S3]. We confirm a RS like behaviour of \( m_s(h_s) \) at \( T = 0 \) also with DMRG as shown in Fig. 2b in main text.

**Figure S4.** (Color online). Finite size dependence of the \( T = 0 \) staggered magnetization \( m_s \) as a function of staggered field \( h_s \), as calculated for \( L = 100, 200, 400, 800 \). It is evident that by increasing the system size \( L \), more reliable or converged results are obtained for smaller \( h_s \). Black line represents semi-analytical solution proposed by Schulz [S4] (see Eq. (6) with corresponding discussion in the main text).

**Figure S5.** (Color online). System size (\( L \)) dependence of \( \chi_\pi \) at finite temperature \( T \), as calculated for different \( L = 20, 40, 60, 80 \) and (a) \( \delta J = 0 \) and (b) \( \delta J = 0.6 \). It is clear that in the shown regime (\( T > 0.1, 0.05 \)) results have converged with system size. Black solid line on panel (a) represents a fit to analytical solution [S7–S9], \( \chi_\pi = a \sqrt{\log(b/T)/T} \) [Eq. (4) in the main text]. Black solid line on panel (b) represents \( \chi_\pi \) calculated from dynamical spin susceptibility with finite-temperature Lanczos method (\( L = 24, 100 \) realizations).
Figure S6. (Color online). (a,b) Temperature dependence of $m_s/h_s$ for different values of $h_s$, as calculated for $L = 80$ sites and (a) $\delta J = 0$ and (b) $\delta J = 0.6$. It is evident that lowering $T$ requires smaller field $h_s$ to reach saturation of $m_s/h_s$ and therefore the linear regime with $m_s = \chi \pi h_s$. (c,d) Field dependence of $m_s/h_s$ for different values of temperature $T$, as calculated for $L = 80$ sites ($T = 0$ result is calculated with $L = 800$ sites) and for $\delta J = 0$ (c) and $\delta J = 0.6$ (d). It is clear that at sufficiently low $h_s$, at given $T$ the values of $m_s/h_s$ are saturating. E.g., for $\delta J = 0$ ($\delta J = 0.6$) and $T = 0.1$ ($T = 0.05$) the lowest presented fields have the same value. This is however not the case for lower $T = 0.075$ ($T = 0.025$), where lower fields would be needed.

Figure S7. (Color online). Staggered field $h_s$ dependence of staggered magnetization $m_s$ for several temperatures $T$ and (a) $\delta J = 0$ and (b) $\delta J = 0.6$. Calculated for $L = 40$ sites ($T = 0$ result is calculated with $L = 800$ sites).

[S1] C. Dasgupta and S.-k. Ma, Phys. Rev. B 22, 1305 (1980).
[S2] A. Joshi and K. Yang, Phys. Rev. B 67, 174403 (2003).
[S3] D. Fisher, Phys. Rev. B 50, 3799 (1994).
[S4] H. J. Schulz, Phys. Rev. Lett. 77, 2790 (1996).
[S5] J. Kokalj and P. Prelovšek, Phys. Rev. B 80, 205117 (2009).
[S6] J. Herby, J. Kokalj, and P. Prelovšek, Phys. Rev. Lett. 111, 147203 (2013).
[S7] T. Giamarchi and H. Schulz, Phys. Rev. B 39, 4620 (1989).
[S8] R. R. P. Singh, M. E. Fisher, and R. Shankar, Phys. Rev. B 39, 2562 (1989).
[S9] I. Affleck, D. Gepner, T. Ziman, and H. J. Schulz, J. Phys. A: Math. Gen. 22, 511 (1989).