**Thermodynamic Spin Glass Phase Induced by Weak Random Exchange Disorder in a Classical Spin Liquid: the Case of the Pyrochlore Heisenberg Antiferromagnet**

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Most magnetic materials develop long range magnetic order when the temperature is sufficiently low. There are, however, two prominent exceptions: spin liquid (SL) and spin glass (SG) systems. Both of them are commonly found among frustrated systems where the ordering tendency is reduced. The SL usually occurs in geometrically frustrated systems, whereas the SG arises in random frustrated systems due to competing random antiferromagnetic (AFM) and ferromagnetic (FM) couplings.

The Y$_2$Mo$_2$O$_7$ pyrochlore Heisenberg antiferromagnet, possibly with some form of weak disorder, whose origin remains both mysterious and controversial, does not fall in the category of conventional SG materials. The Heisenberg AFM model on the three-dimensional pyrochlore lattice of corner sharing tetrahedra is a classical spin liquid (CSL) with macroscopically degenerate ground states which satisfy the zero net magnetic moment ($\Phi_0 = 0$) constraint on each tetrahedron. This constraint leads to a gauge theory description of the CSL and the prediction of a power-law decaying spin-spin correlation function of “dipolar” form. Thus, a clean Heisenberg AFM model on the pyrochlore lattice does not display a SG state. However, the macroscopic degeneracy in this model opens up the interesting possibility that weak random disorder in the spin-exchange interactions, so small that no competing AFM-FM coupling is present in the bare Hamiltonian, may be sufficient to induce a SG phase at nonzero temperature. The main question that we address in this paper is whether dense random weak disorder in the AFM exchange can induce a thermodynamic SG phase from a CSL as a case study of what may be occurring in Y$_2$Mo$_2$O$_7$.

We study the Hamiltonian $\mathcal{H}$ defined on a pyrochlore lattice first proposed by Bellier-Castella et al.:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{dis}},$$  
(1)

with $\mathcal{H}_0 = J_0 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$ and $\mathcal{H}_{\text{dis}} = \sum_{\langle i,j \rangle} J_{ij}^{\text{dis}} \mathbf{S}_i \cdot \mathbf{S}_j$, where $\{\mathbf{S}_i\}$ are three component unit vectors and summations are over nearest neighbors. $\mathcal{H}_0$ is the usual pyrochlore Heisenberg AFM model which displays on its own a CSL while $\mathcal{H}_{\text{dis}}$ describes the random disorder which mimics the situation in Y$_2$Mo$_2$O$_7$. Weak disorder here means $|J_{ij}^{\text{dis}}| \ll J_0$. We set the Boltzmann constant $k_B = 1$, and also set $J_0 = 1$ which serves as overall energy scale. The bond disorder is uniformly distributed in the range $J_{ij}^{\text{dis}} = [-W, W]$ with $W = J_0/10$ used in the calculations. We refer to $\mathcal{H}$ in Eq.(1) as the BGHM model.

The original work of Bellier-Castella et al. suggested, on the basis of measurements of the SG overlap parameter, that glassy behavior in $\mathcal{H}$ sets in at a temperature roughly the same as $W^{10}$. In more recent studies, Saunders and Chalker and Andreanov et al. computed the SG correlation function and SG susceptibility. Based on numerical data and analytic arguments, the authors of Ref. 12 suggested that there exists a thermodynamic SG transition at a nonzero temperature for arbitrary small but nonzero $W$.

SG simulations techniques have significantly improved over the past ten years or so. Recent extensive Monte Carlo simulations employing these improvements have led to the revision of the old belief that the lower critical dimension, $d_l$, of the Edwards-Anderson (EA) isotropic Heisenberg SG model is above three. There is no rigorous analytic approach to determine the $d_l$ of the BGHM model and the previous MC simulations do not come close to the computational standard of recent studies of the EA Heisenberg SG. Thus, the analytic arguments and numerical data at hand can hardly provide convincing evidence for a thermodynamic SG phase in $\mathcal{H}$. The BGHM model, with its underlying CSL state in the disorder-free regime, as well as its broad relevance to the SG behavior observed in numerous geometrically frustrated magnetic materials, make it a model of fundamental significance in the field of frustrated magnetism. It is therefore important to carefully assess whether $\mathcal{H}$ sustains a thermodynamic SG phase at nonzero temperature in $\mathcal{H}$.

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SG phase at nonzero temperature, and to reach such a conclusion on the basis of numerics that approach the standard of SG simulations of EA models.

We first summarize the details of our MC simulations. A 16 site cubic unit cell for the pyrochlore lattice is used for generating cubic simulation cells with \( N = 16L^3 \) spins with \( L = 4, 6, 8 \). One Metropolis sweep, 2L over-relaxation sweeps and one parallel tempering sweep\(^{22}\) is defined as one elementary MC step (MCS). The temperatures explored for each simulation are \( T(\kappa) = T_{\text{min}}(\kappa)^{-1} \) where \( T_{\text{min}} \) is the lowest temperature considered and \( N \) is the number of thermal replicas. Thus the highest temperature is \( T_{\text{max}} = T_{\text{min}}(\kappa)^{N_T-1} \) and \( \kappa = (\kappa_T-1)/T_{\text{max}}/T_{\text{min}} \). The error bars are sample-to-sample fluctuations calculated via the jackknife method. Table I lists the parameters used in our MC simulations.

### Table I: Parameters of the Monte Carlo simulations.

| \( L \) | \( T_{\text{min}} \) | \( T_{\text{max}} \) | \( N_T \) | \( N_{\text{MCS}}^\text{MC} \) | \( N_{\text{MCS}}^\text{measurement} \) | \( N_{\text{samples}} \) |
|------|----------------|----------------|-------------|----------------|-----------------|----------------|
| 4    | 0.012 0.028   | 24 2 \( \times 10^4 \) | 2 \( \times 10^3 \) | 1750           |
| 6    | 0.012 0.028   | 30 4 \( \times 10^4 \) | 2 \( \times 10^4 \) | 1763           |
| 8    | 0.012 0.028   | 46 8 \( \times 10^4 \) | 2 \( \times 10^4 \) | 1613           |

To characterize a putative SG phase, we use a parameter defined as the overlap between two thermal replicas 

\[
\chi_{SG}(k) = N \sum_{\mu,\nu=x,y,z} (|\langle S^{(1)}_{\mu,\nu} S^{(2)}_{\mu,\nu} \rangle|^{2} / 2) \]

where \( S^{(1)}_{\mu,\nu} \) and \( S^{(2)}_{\mu,\nu} \) are the spin components for replicas (1) and (2), respectively. It has been proposed that there is no SG transition in isotropic Heisenberg SG systems but that, instead, the freezing is in the chiral sector. Latest simulations\(^{17,21}\) suggest that both chiral glass (CG) and SG transitions occur at finite temperature, but there is no consensus whether the critical temperature \( T_{CG} \) is higher or equal to that of the SG \( T_{SG} \). Since \( H \) has isotropic Heisenberg spins, there is no obvious reason to exclude the possibility of a CG transition. To monitor CG correlations, we consider two chirality parameters. The first one is defined along bonds,

\[
q_{CG1}(k) = \frac{1}{3N} \sum_{i=1}^{N} \sum_{\delta=\pm} S^{(1)}_{\iota,\delta} S^{(2)}_{\iota,\delta} \exp(i k \cdot r_i),
\]

where \( S^{(1)}_{\iota,\delta} \) and \( S^{(2)}_{\iota,\delta} \) are the spin components for replicas (1) and (2), respectively. It has been proposed that there is no SG transition in isotropic Heisenberg SG systems but that, instead, the freezing is in the chiral sector. Latest simulations\(^{17,21}\) suggest that both chiral glass (CG) and SG transitions occur at finite temperature, but there is no consensus whether the critical temperature \( T_{CG} \) is higher or equal to that of the SG \( T_{SG} \). Since \( H \) has isotropic Heisenberg spins, there is no obvious reason to exclude the possibility of a CG transition. To monitor CG correlations, we consider two chirality parameters. The first one is defined along bonds,

\[
q_{CG2}(k) = \frac{1}{2N} \sum_{\omega=1}^{2N} \kappa^{(1)}_{\omega,\omega} \kappa^{(2)}_{\omega,\omega} \exp(i k \cdot r_\omega),
\]

where \( \kappa^{(1)}_{\omega,\omega} = S^{(1)}_{\omega,\omega} \cdot (S^{(1)}_{\omega,\omega} \times S^{(1)}_{\omega,\omega}) \); \( \omega, a, (\omega, b), (\omega, c) \) are the indices for the three sites of the triangular face \( \omega \) and \( r_\omega = (r_\omega, a + r_\omega, b + r_\omega, c)/3 \). The corresponding susceptibilities are obtained from the order parameters,

\[
\chi_{CG1}(k) = 3N \langle |\chi_{CG1}(k)|^2 \rangle / \chi_1^2, \quad \chi_{CG2}(k) = 2N \langle |\chi_{CG2}(k)|^2 \rangle / \chi_2^2,
\]

where \( \langle \ldots \rangle \) and \( \langle \ldots \rangle \) denote the thermal average and disorder average, respectively. As the local chirality variables are not fixed to be unity as is the case of the spin variables, the CG susceptibility is normalized by \( \chi_1^2 \) whereas the SG susceptibility is normalized by \( \chi_2^2 \), and, similarly, the CG2 susceptibility is normalized by \( \chi_2^2 \), see Appendix).

Assuming that the susceptibilities follow an Ornstein-Zernike form, the correlation lengths \( \xi_{SG} \) and \( \xi_{CG1,CG2} \) can be determined via \( \xi_{\gamma}(L) = \frac{1}{2 \chi_{\gamma}(k=2)} \left( \frac{\chi_{0}(0)}{\chi_{0}(k)} - 1 \right)^{1/2} \), where \( \gamma \equiv SG, CG1 \) or \( CG2 \) and \( k = 2 \pi \delta / L \) is one of the smallest wave vectors for system size \( L \). The \( \xi_{\gamma}'s \) divided by \( L \) should be scale invariant at their respective critical point. The crossing of \( \xi_{\gamma}(L) \) is therefore a sensitive criterion to test for a glass transition. The correlation lengths and susceptibilities should finite-size scale as \( \xi_{\gamma}(L)/L = X[(T - T_{\gamma})L^{1/\nu_{\gamma}}] \) and \( \chi_{\gamma}(T, L)L^{1/\nu_{\gamma} - 2} = Y[(T - T_{\gamma})L^{1/\nu_{\gamma}}] \), respectively. To check that thermodynamic equilibrium was reached, we verified that \( \xi_{\gamma} \) becomes independent of simulation time for the largest system size and lowest temperature considered.

We first present the SG and CG correlation lengths in Fig. 1. It is fairly clear from these results that the SG and CG correlation lengths for different system sizes tend to cross in a narrow range of temperatures compatible with a nonzero critical temperature for both SG and CG \( (T_{\gamma} \approx 0.016) \).

We then employ a scaling scheme which assumes that the correlation lengths finite-size scale as the scaling function \( X \) given above and fit the data in the temperature range \( (0.012 \leq T \leq 0.020) \) by parametrizing \( X \) as polynomials \( X(z) = \sum_{m=0,1,2,5} c_m (z - z_0)^m \). The merit function \( \Delta, \Delta = \sum_{MC \text{ data}} |X(z)/(\xi_{\gamma}, L, L^{-1}) - 1|^{2} \), is minimized numerically to obtain the coefficients \( c_m, z_0 \), critical temperature \( T_{\gamma} \) and exponent \( \nu_{\gamma} \). Figure 2 shows \( \xi_{\gamma}(L)/L \) versus the scaling parameter \( z \equiv (T - T_{\gamma})L^{1/\nu_{\gamma}} \) \( (T_{\gamma} \) and \( \nu_{\gamma} \) are listed in the caption of Fig. 2). The scaling exponents determined both for the SG and CG are far from those of the 3D EA Ising model obtained from correlation length scaling \( (\nu_{SG} \approx 2.44 \pm 0.18) \), but roughly comparable (within \( \sim 20\% \)) to those of the 3D EA Heisenberg model\(^{17,21,24}\). In the above scaling analyses, we assumed that there is a common crossing point for all system sizes. Realistically, the critical temperature obtained this way for fair system sizes \( (N \leq 8192) \) should represent an upper bound for the true critical temperature in the thermodynamic limit. For example, in the latest simulations of the 3D EA Heisenberg model, it was found that scaling corre-
data collapse to fit the susceptibilities into the scaling function, \( \chi \), in order to determine \( T_g, \nu_g \) and \( \eta_g \). Figure 2 shows \( \chi(T, L) L^{\nu_g - 2} \) versus the scaling parameter \( z = (T - T_g) L^{1/\nu_g} \). (The \( T_g, \nu_g \), and \( \eta_g \) are listed in the caption of Fig. 2.) The \( \eta_g \) and \( \nu_g \) values are again fairly comparable with those obtained in the latest studies of the 3D EA Heisenberg model\(^{17,21,23} \), providing evidence for a common SG universality class for the BGHM model.
and the 3D EA Heisenberg model.

A recent study aimed at describing the SG in $H$ assumes that the power-law correlation of the CSL is maintained despite the random disorder and thus the spins can be thought of as interacting via an effective projected interaction matrix of a long range “dipolar” form as a consequence of the zero net magnetic moment ($\Phi_{t=0}$) condition on each tetrahedron. To investigate this description, we calculate the average tetrahedra moment, $\Phi$, vs temperature, $T$.

![Figure 3](Figure 3: (Color online). Left: Correlation length crossing temperatures for SG, CG1 and CG2 for different pairs of system sizes $(L_1, L_2)$. The vertical axes are the crossing temperatures and the horizontal axis is the inverse of the average system sizes given by $1/L_{ave} = 2/(L_1 + L_2)$. The lines are guides for linear extrapolations to the limit $L_{ave} \rightarrow \infty$, that is assuming $T_0^\xi(L_1, L_2) = T_0 \propto L_{ave}^{-\theta}$ with $\theta = 1$. Right: The average tetrahedra moment, $\Phi$, vs temperature, $T$.

of $Y_2Mo_2O_7$ is not due to weak random disorder as in $H$, but rather to very strong effective disorder. One plausible scenario is that perturbations beyond nearest-neighbor Heisenberg exchange $J_0$ disrupt the perfect degeneracy of the CSL phase and induce short range AFM order above $T_g$, as observed in a neutron scattering study. If the growth of AFM order is forestalled due to some form of random disorder, the SG behavior of $Y_2Mo_2O_7$ should likely be described in terms of a “cluster-glass” model.

In conclusion, our MC simulations of the BGHM model of Eq. (1) provide compelling evidence for a thermodynamic SG phase induced by weak random disorder in a classical spin liquid of a highly frustrated system. From our work, it appears very likely that the SG behavior in $Y_2Mo_2O_7$ is not due to weak and dense random disorder but rather via an effective strong disorder whose microscopic origin requires further investigation.

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Appendix A: Spin and Chiral Glass Susceptibilities

This appendix discusses the details of the definitions and calculations of the spin glass (SG) and different chiral glass (CG) overlap parameters — CG1 defined along the bonds and CG2 defined on the triangular faces of the tetrahedra that from a pyrochlore lattice. In addition, a table for critical exponents obtained from previous Monte Carlo studies of the three-dimensional (3D) Edwards-Anderson (EA) Heisenberg SG model is provided for comparison with the critical exponents obtained for the BGHM model studied in this paper.

The spin glass (SG) overlap is defined as the overlap between two thermal replicas with the same realization of random couplings $\{J_{ij}^{\text{dis}}\}$,

$$q_{SG}^{\mu,\nu}(k) = \frac{1}{N} \sum_{i=1}^{N} S^{(1)}_{i,\mu} S^{(2)}_{i,\nu} \exp(i k \cdot r_i), \quad (A1)$$

where $S^{(1)}_{i,\mu}$ and $S^{(2)}_{i,\nu}$ are the spin components of the two replicas. For Ising spins, this is the usual parameter used to monitor the spin freezing. The situation is, however, more complicated for Heisenberg spins.

It has been proposed that there is no SG transition in isotropic Heisenberg SG systems but, instead, that the freezing is in the chiral sector. There have been many investigations on the spin-chirality coupling/decoupling scenario for the 3D EA Heisenberg model, but there is so far no consensus, while recent studies tend to agree that both the critical temperature for SG ($T_{SG}$) and chiral glass ($T_{CG}$) are non-zero, whether $T_{CG} = T_{SO}$ or $T_{CG} > T_{SO}$ is still under active investigations.

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debate. Since the model we study possesses spins with isotropic Heisenberg exchange, there is no a priori reason to rule out the possibility of a chiral glass (CG) transition.

To monitor the CG we probe two different chirality variables. The first one is defined along the bonds. This is a generalization of the definition of chirality variables for the 3D EA Heisenberg model on a simple cubic lattice. Similarly to the simple cubic lattice, three are axes passing thorough each site. As three spins are needed to define the chirality, the natural choice is to pick a spin and its two nearest neighbors spins along one of the axes to define a chirality variables. As there are three axes passing thorough each site, therefore there are three chirality variables for each site. We denote this definition of chirality overlap as CG1.

\[ q_{CG1}(k) = \frac{1}{3N} \sum_{i=1}^{N} \sum_{\delta} \nabla_{\delta i} \cdot \hat{\epsilon}_{i,1} \cdot \hat{\epsilon}_{i,2} \cdot \hat{\epsilon}_{i,3} \exp(i \cdot \mathbf{r}_i) \]

where \( \nabla_{\delta} = \hat{\epsilon}_{i,\delta} \) (\( \hat{\epsilon}_{i,\delta} \) are vectors pointing from site \( i \), to its three nearest neighbor sites in the same tetrahedron, see Fig. 4) \( (i, \delta) \) are the indices for the three sites lying along \( \delta \). The normalization factor \( 1/3N \) is introduced to account for the \( 3N \) chirality variables for this definition because of the three chirality variables at each site. We note that a slightly different definition, which treats the CG overlap as a three components object has been used in a study of 3D EA Heisenberg model\(^{22} \).

The second chirality overlap parameter is defined on the triangular faces of the tetrahedra. The pyrochlore lattice is composed of corner-sharing tetrahedra, and each tetrahedron has 4 triangular faces. Therefore, another natural choice is to define the chirality variables on each face of the tetrahedra. In a lattice with \( N \) sites, there are \( N/2 \) tetrahedra, and each tetrahedron has 4 faces. Therefore, there are in total \( 2N \) chirality variables within this definition. We denote this definition of chirality as CG2, with:

\[ q_{CG2}(k) = \frac{1}{2N} \sum_{\omega=1}^{2N} \kappa^{(1)}_{\omega}(k) \kappa^{(2)}_{\omega}(k) \exp(i \cdot \mathbf{r}_\omega), \]

where \( \kappa^{(1)}_{\omega} = \mathbf{S}_{\omega,a} \cdot (\mathbf{S}_{\omega,b} \times \mathbf{S}_{\omega,c}) \), \( (\omega, a), (\omega, b), (\omega, c) \) are the indices for the three sites of the triangular face \( \omega \) and \( \hat{\mathbf{r}}_\omega = (\hat{\mathbf{r}}_{\omega,a} + \hat{\mathbf{r}}_{\omega,b} + \hat{\mathbf{r}}_{\omega,c})/3 \). The normalization factor \( 1/2N \) is introduced to account for the \( 2N \) chirality variables for this definition since there are \( 2N \) triangular faces.

The corresponding susceptibilities are obtained from the above order parameters,

\[ \chi_{SG}(k) = N \sum_{\mu,\nu=1}^{2N} \langle |\rho^{\mu\nu}_{SG}(k)|^2 \rangle; \]

\[ \chi_{CG1}(k) = 3N \langle |q_{CG1}(k)|^2 \rangle / \chi_1^4, \]

\[ \chi_{CG2}(k) = 2N \langle |q_{CG2}(k)|^2 \rangle / \chi_2^4, \]

where \( \langle \ldots \rangle \) and \( \langle \ldots \rangle \) denote the thermal average and disorder average respectively. As the local chirality variables are not fixed to be unity in contrast of the spin variables\(^{19,20,35} \), the CG1 susceptibility is normalized by \( \chi_1^4 = (\chi_1^2)^2 \), where

\[ \chi_1^2 = \frac{1}{3N} \sum_{i=1}^{N} \sum_{\delta} \langle |\kappa^{(1)}_{1,\delta}|^2 \rangle, \]

and the CG2 susceptibility is normalized by \( \chi_2^4 = (\chi_2^2)^2 \), where

\[ \chi_2^2 = \frac{1}{2N} \sum_{\omega=1}^{2N} \langle |\kappa^{(2)}_{2,\omega}|^2 \rangle. \]

The exponents obtained for the BGHM model are fairly comparable with that of the 3D EA Heisenberg spin glass model\(^{22} \). For comparison of our results with the 3D EA Heisenberg model, we compile a selection of critical temperatures and critical exponents in Table II For the 3D EA Ising model, see Table I in Ref. [15].
| reference                        | randomness type | $T_{SG}$ | $\nu_{SG}$ | $\eta_{SG}$ | $T_{CG}$ | $\nu_{CG}$ | $\eta_{CG}$ |
|---------------------------------|-----------------|----------|-----------|-------------|----------|-----------|-------------|
| Kawamura** (1998)               | Gaussian        | NA       | NA        | NA          | 0.157 ± 0.01 | NA        | NA          |
| Hukushima and Kawamura** (2000) | Gaussian        | NA       | NA        | NA          | 0.160 ± 0.005 | 1.2       | 0.8         |
| Endoh, et al.** (2001)          | Bimodal         | 0.19 ± 0.02 | NA        | NA          | NA        | NA        | NA          |
| Matsubara, et al.** (2001)      | Bimodal         | 0.18     | NA        | NA          | 0.22 ± 0.01 | NA        | NA          |
| Nakamura and Endoh** (2002)     | Bimodal         | 0.21 ± 0.01 | 1.1 ± 0.2 | 0.27#       | NA        | NA        | NA          |
| Lee and Young** (2003)          | Gaussian        | 0.16 ± 0.02 | 1.1 ± 0.2 | NA          | $T_{CG} = T_{SG}$ | 1.3 ± 0.3 | NA          |
| Nakamura, et al.** (2003)       | Bimodal         | 0.20 ± 0.02 | 0.8 ± 0.2 | $-0.375$#   | NA        | NA        | NA          |
| Hukushima and Kawamura** (2005) | Bimodal         | NA       | NA        | NA          | 0.19 ± 0.01 | 1.3 ± 0.2 | 0.8 ± 0.2   |
| Viet and Kawamura** (2009)      | Gaussian        | 0.125 ± 0.012 | NA       | $\zeta$ ± 0.30 | 0.143 ± 0.003 | 1.4 ± 0.2 | 0.6 ± 0.2   |
| Fernandez, et al.** (2009)      | Gaussian        | 0.120 ± 0.004 | 1.49 ± 0.13 | $-0.19 ± 0.02$ * | $T_{CG} = T_{SG}$ | 1.30 ± 0.08 * | 0.56 ± 0.04 * |
| This work ($\xi/L$ scaling)     | pyrochlore      | 0.0157   | 1.037     | NA          | 0.0153(CG1) | 1.585(CG1) | NA          |
|                                | ±0.1L₀ uniform  |          |          |             | 0.0161(CG2) | 1.408(CG2) | NA          |
| This work ($\chi$ scaling)      | pyrochlore      | 0.0126   | 1.024     | $-0.292$    | 0.0134(CG1) | 1.439(CG1) | 0.621(CG1)  |
|                                | ±0.1L₀ uniform  |          |          |             | 0.0139(CG2) | 1.155(CG2) | 0.698(CG2)  |

TABLE II: Selection of critical temperatures and exponents of the three-dimensional Edwards-Anderson Heisenberg model for the simple cubic lattice. For the Edwards-Anderson Ising model see Table I in Ref. [15]. NA is a shorthand for not available. The last two rows are the estimates from this work. The variance of the random coupling distribution is 1 for all the studies on the three-dimensional Edwards-Anderson model listed in this table, and the variance of the model we study is $1/3 \times 10^{-2}$. (The variance is defined as $\int (x - \bar{x})^2 P(x) dx$, where $P(x)$ is the distribution function and $\bar{x} = \int x P(x) dx$.) The entries with “#” are not quoted in the original paper but are estimated via the relation $\gamma = (2 - \eta)\nu$. The entries with “*” are from the quotient method for scaling analysis of $L = 24$ and $L = 48$ systems on a simple cubic lattice.

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