Magnetic and quadrupolar order in a one-dimensional ferromagnet with cubic crystal-field anisotropy

M. Dudziński†, G. Fáth‡, and J. Sznajd†
†Institute for Low Temperature and Structure Research, Polish Academy of Sciences, P. O. Box 1410, 50-950 Wroclaw 2, Poland
‡Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, England

The zero temperature phase diagram of a one-dimensional $S = 2$ Heisenberg ferromagnet with single-ion cubic anisotropy is studied numerically using the density-matrix renormalization group method. Evidence is found that although the model does not involve quadrupolar couplings, there is a purely quadrupolar phase for large values of the anisotropy. The phase transition between the magnetic and quadrupolar phases is continuous and it seems to be characterized by Ising critical exponents.

I. INTRODUCTION

It is well-known that crystal fields present in real materials can influence their magnetic behavior considerably. In this paper we study a ferromagnetic insulator, with localized interacting magnetic moments, which are also subject to a crystal field of cubic symmetry. The finite-temperature critical phenomena in these systems have been investigated previously within the Landau-Ginzburg-Wilson theory and the cubic universality class and its critical exponents have been analyzed. However, in such phenomenological theories, the form of the order parameter is simply postulated, using mainly symmetry arguments. To see whether such an order really develops in the low-temperature regime one must start with the microscopic quantum Hamiltonian.

In terms of a quantum spin Hamiltonian, the magnetic moments are described by spin operators, and the crystal field takes the form of a single-site term. The lowest value of $S$ for which a cubic anisotropy term has any effect is $S = 2$. Using this value our model Hamiltonian takes the form

$$H = -\sum_{\langle ij \rangle} S_i S_j - D \sum_i [(S_i^x)^4 + (S_i^y)^4 + (S_i^z)^4],$$

with $S_i^\alpha$ ($\alpha = x, y, z$) denoting spin operators for $S = 2$ at lattice site $i$. The first term is the isotropic, ferromagnetic exchange interaction between nearest neighbors, and the second term represents the crystal field anisotropy; for $S = 2$ this is the most general single-site operator with cubic symmetry. The exchange term alone produces ferromagnetic order in the low-temperature phase; the question we want to address is how this order is affected by the cubic crystal field.

The above Hamiltonian has been treated within the mean-field approximation (MFA) by many authors. It has been found that the sign of the crystal-field constant $D$ defines the easy axes of the magnetic ordering at low temperatures. For $D = 0$ the system is isotropic and the spontaneous magnetization can lie along any direction; for $D < 0$ the cube diagonals [111] are preferred; for $0 < D < (4z)/3$, where $z$ is the number of nearest neighbors, the directions parallel with the cube edges [100] are chosen. Moreover, the MFA predicts that for $D > (4z)/3$ the system is disordered, irrespective of the temperature.

Recently, two of us (M.D. and J.S.) have investigated the above model applying the Raleyigh-Schrödinger perturbation theory in the limit $D \to \infty$. This calculation is valid for large $D$, and the following picture emerges from it. In agreement with the MFA, for large $D$, there is no magnetic order at any temperature, i.e., the averages of the spin operators vanish:

$$\langle S_i^\alpha \rangle = 0, \quad \text{for} \quad \alpha = x, y, z.$$  

However, in contrast to the MFA prediction, the symmetry may still be spontaneously broken by the appearance of a purely quadrupolar order, described by the averages

$$\langle (S_i^x)^2 \rangle \neq \langle (S_i^y)^2 \rangle = \langle (S_i^z)^2 \rangle.$$  

(One may interchange $x$, $y$, and $z$ in the above relation.) This purely quadrupolar phase was predicted to exist in the model on a two-dimensional lattice at least at zero temperature. In three dimensions and in higher dimensions it should be present both in the ground state and at finite (but low) temperatures. We suppose, that the quadrupolar phase also exists in one dimension at zero temperature (note that the broken symmetry is not continuous but discrete), although in this case the perturbation-theory arguments are weaker.

The result that there may appear a purely quadrupolar order in the model in Eq. (1), which contains only the bilinear, “magnetic” exchange and the single-site term, is rather surprising. Usually, higher order exchanges are responsible for quadrupolar ordering. The appearance of quadrupolar order in the present case is entirely a quantum effect; one would not obtain purely quadrupolar order by simply replacing the spin operators in Eq. (1) with classical vectors, and then seeking the configuration...
with minimum energy. The result is also of methodological interest. It is easy to understand that one cannot get purely quadrupolar order in this model by the MFA: only magnetization can act as a mean field when the exchange term contains only first powers of the spin operators. Therefore, if the purely quadrupolar phase exists, we have the example that, in contrast with the general belief, the MFA may give an answer which is qualitatively wrong in high dimensions.

Unfortunately, there is no fully reliable method which would enable us to investigate the model in two or three dimensions, for general $D$. Therefore, the aim of the present paper is to check and confirm the existence of the quadrupolar phases. We also attach an Appendix in which numerical predictions, obtained using identical techniques, are confronted with the exact results in the test case of the 1D Ising model in a transverse field.

We discuss how long-range order can be detected using the DMRG in Sec. [V]. Section [V] is devoted to the concrete numerical results, while Sec. V is a brief discussion of our conclusions. We also present an Appendix in which numerical predictions, obtained using identical techniques, are confronted with exact results in the test case of the 1D Ising model in a transverse field.

II. PREDICTIONS FROM MFA AND PERTURBATION THEORY

Let us first list the eigenvalues $e_k$ and eigenstates $|\psi_k\rangle$ of the single-site term in the Hamiltonian in Eq. (1):

\[e_1 = -24D \quad |\psi_1\rangle = \frac{1}{\sqrt{2}}(|2\rangle + | -2\rangle),\]
\[e_2 = -24D \quad |\psi_2\rangle = |0\rangle,\]
\[e_3 = -18D \quad |\psi_3\rangle = |1\rangle,\]
\[e_4 = -18D \quad |\psi_4\rangle = \frac{1}{\sqrt{2}}(|2\rangle - | -2\rangle),\]
\[e_5 = -18D \quad |\psi_5\rangle = -| -1\rangle,\]

where by $|2\rangle, |1\rangle, \ldots, | -2\rangle$ we denote eigenstates of $S^z$. The five states are split into a doublet and a triplet, and the doublet energy is lower for $D > 0$. The doublet is nonmagnetic, i.e.,

\[\langle \psi_i | S^\alpha | \psi_j \rangle = 0,\]

for any $i, j = 1, 2$ and $\alpha = x, y, z$. Clearly, the crystal field with $D > 0$ opposes the magnetic ordering, since the nonmagnetic states are preferred in the total wave function of the system.

For $D = 0$ the model is isotropic and possesses the classical, ferromagnetic ground state with saturated magnetization. As mentioned in the Introduction, the MFA predicts that the magnetic phase exists for $0 < D < 8/3$ ($z = 2$ for the chain), and that the directions of the cube edges $[100]$ are the easy axes in this phase. According to the MFA, the ground-state magnetization decreases with $D$ and disappears continuously at $D = 8/3$. The MFA result that the magnetic order vanishes at some finite $D$ is a strong argument that this is indeed the case, since the MFA usually overestimates the tendency towards ordering. Nevertheless, the actual value of $D$ at which the magnetic phase ends may be much smaller than $8/3$ (for example, in the Ising chain in a transverse field the MFA critical field is two times larger than the exact critical field). Whether the actual phase transition is continuous or discontinuous is also a matter of question. We believe that the easy axes in the magnetic phase are given correctly by the MFA. This approximation neglects the quantum fluctuations in the ground state, but it seems very unlikely that these could change the easy-axis directions, especially in the vicinity of $D = 0$, where the magnetization is almost saturated and the quantum fluctuations are small.

Considering the large-$D$ limit now, we make use of the perturbation theory developed in Ref. [4]. Since the two nonmagnetic states $|\psi_1\rangle$ and $|\psi_2\rangle$ dominate for large $D$, the model reduces to an effective two-state Hamiltonian. Restricting our attention to the one-dimensional situation from now on, the effective Hamiltonian $H_{\text{eff}}$ can be written in terms of Pauli matrices $\sigma^\alpha_i$ acting on the states $|\psi_1\rangle$ and $|\psi_2\rangle$, and has the form

\[H_{\text{eff}} = -\tilde{J}_1 \sum_i (\sigma^x_i \sigma^x_{i+1} + \sigma^z_i \sigma^z_{i+1}) - \tilde{J}_2 \sum_i (\sigma^y_i \sigma^y_{i+1}) - \tilde{J}_3 \sum_i (\sigma^x_i \sigma^z_{i+1} + \sigma^z_i \sigma^z_{i+1}) - \tilde{J}_4 \sum_i (\sigma^x_i \sigma^z_{i+1} \sigma^z_{i+2} - \sigma^z_i \sigma^z_{i+1} \sigma^z_{i+2} - \sigma^z_i \sigma^z_{i+1} \sigma^z_{i+2}) + O\left(\frac{1}{D^4}\right),\]

with

\[\tilde{J}_1 = \frac{1}{2D} - \frac{1}{12D^2} + \frac{1}{72D^3} + O\left(\frac{1}{D^4}\right),\]
\[\tilde{J}_2 = \frac{1}{8D^2} + \frac{1}{32D^3} + O\left(\frac{1}{D^4}\right),\]
\[\tilde{J}_3 = \frac{1}{18D^3} + O\left(\frac{1}{D^4}\right),\]
\[\tilde{J}_4 = \frac{1}{24D^3} + O\left(\frac{1}{D^4}\right).\]

(The exchange coupling constant $J$ from Ref. [4] is set to 1 in the present work.) The quadrupole operators $[(S^\alpha_i)^2 - 2]$ of the original model in Eq. (1) turn out to
be represented by linear combinations of Pauli matrices as

\[
\begin{align*}
(S^z_i)^2 - 2 & \quad \mapsto \sqrt{3} \sigma_i^1 - \sigma_i^2 + \mathcal{O}(1/D^2), \\
([S^y_i])^2 - 2 & \quad \mapsto -\sqrt{3} \sigma_i^1 - \sigma_i^2 + \mathcal{O}(1/D^2), \\
([S^x_i])^2 - 2 & \quad \mapsto 2 \sigma_i^2 + \mathcal{O}(1/D^2).
\end{align*}
\]

(5)

The analogous representations of \(S_0^\alpha\) and \((S_0^\alpha S_0^\beta + S_0^\beta S_0^\alpha)\), where \(\alpha \neq \beta\), vanish up to \(\mathcal{O}(1/D^3)\).

In the lowest order in \(1/D\), only \(J_1\) is present in the effective Hamiltonian, and \(H_{\text{eff}}\) describes the planar model with continuous SO(2) symmetry in the \(XZ\) plane. While this model, conventionally written as the XY model, is ordered at zero temperature in two dimensions and above, in one dimension it is known to be critical due to enhanced quantum fluctuations.

The in-plane correlation function

\[\langle \sigma_i^x \sigma_i^x \rangle = \langle \sigma_i^y \sigma_i^{y+1} \rangle\]

decays asymptotically as a power law \(1/\sqrt{l}\), where \(l\) is the distance between the two sites. Hence, on the basis of Eq. (3), we argue that for \(D \to \infty\) a critical state, in which the quadrupolar correlation function

\[\langle ([S_0^\alpha]^2 - 2) ([S_0^\beta]^2 - 2) \rangle, \quad \alpha = x, y, z,
\]

decays asymptotically as \(1/\sqrt{L}\), is approached.

Taking into account the higher-order terms in \(H_{\text{eff}}\), we obtain the planar, \(XY\)-type model with small perturbations. The three-spin term with the coupling constant \(J_2\) is crucial, because it reduces the symmetry of the effective Hamiltonian from the full rotational symmetry in the \(XZ\) plane to the symmetry of discrete rotations through angles ±(2\(\pi\))/3. In the one-dimensional case the \(J_2\) and \(J_3\) terms are expected to be marginally irrelevant but in the lack of a rigorous study of the off-diagonal three-point symmetry-breaking term with \(J_1\), we can only speculate (and then eventually check numerically) whether the \(\mathcal{O}(1/D^3)\) contributions are relevant or irrelevant. If they are relevant, they can stabilize the “magnetic order” (i.e., the nonvanishing averages of \(\sigma_i^\alpha\)) in the effective model \(H_{\text{eff}}\), since the Mermin-Wagner and Coleman’s theorems do not hold when the symmetry is not continuous but discrete. If this is indeed the case, then Eq. (4) implies that for large \(D\) a quadrupolar order, associated with the operators \(([S_i^\alpha]^2 - 2)\), exists in the original model of Eq. (1) (see Ref. [4] for details). Alternatively, if all the perturbations turn out to be irrelevant, there should be a critical phase for large \(D\).

To summarize the above, we present the relevant (order) parameters to be analyzed as suggested by the MFA and the perturbation theory:

\[m^\alpha = \langle S_0^\alpha \rangle, \quad q^\alpha = \langle (S_0^\alpha)^2 - 2 \rangle. \]

(6)

These ground-state averages are bulk values in the thermodynamic limit. Note, that in a disordered state \(m^\alpha = q^\alpha = 0\) for \(\alpha = x, y, z\). Based on the above analysis we expect that for \(0 < D < D_m\) (where \(D_m\) is probably smaller than the MFA value 8/3) a magnetic phase with

\[m^x \neq 0, \quad m^y = m^z = 0, \quad q^x > q^y = q^z, \]

(7)

exists, while for \(D > D_m\) a purely quadrupolar phase with

\[m^x = m^y = m^z = 0, \quad q^x > q^y = q^z \]

(8)

emerges. (The indices \(x, y, z\) in the above descriptions of the phases may be interchanged.) In both phases the original cubic symmetry of the Hamiltonian is spontaneously broken. Note however that the subgroup remaining invariant is different in the two cases. As discussed above, for large \(D\) a critical (disordered) phase with \(q^\alpha = 0\) cannot be excluded either. In any case, the values of \(q^\alpha\) must go to zero for \(D \to \infty\).

III. THE METHOD

The density-matrix renormalization group (DMRG) method is one of the most reliable numerical techniques to study one-dimensional quantum lattice problems. For a detailed description of the algorithm see Ref. [5]. There are different implementations of the technique; nevertheless it seems that the best accurate results can be obtained by investigating finite systems with open boundary conditions. In this setup the DMRG provides good approximations for the ground state and low-lying excited states of long, but finite quantum chains. This can then be supplemented by a detailed finite size scaling analysis using appropriate scaling assumptions.

Usually for the case of spontaneous symmetry breaking the existence of long-range order is checked by studying appropriate two-point correlation functions \(\langle A_n A_n' \rangle\). The direct observation of one-point functions \(\langle A_n \rangle\) is apparently hindered by the fact that in finite systems, where tunneling is always finite different between different vacua, the ground state is a symmetric combination of all possible ordering directions, and thus naively \(\langle A_n \rangle = 0\). A possible way out, however, is to break the symmetry artificially by a (small) auxiliary field which then forces the system to make a choice between the \(a\ priori\) undetermined directions, without noticeably changing the value of the order parameter. This method has considerable advantages in the DMRG numerics where the measurement of two-point functions is rather awkward due to the open boundary condition.

In our implementation the symmetry-breaking auxiliary fields are only applied to the first and last spins of the open chain. These two \emph{boundary fields} are chosen to be identical, leaving the system symmetric for \(i \to L - i\) reflections (\(L\) is the chain length). Then the Hamiltonian which is actually simulated in the numerical calculations is

\[H \to H - h \cdot (B_1 + B_L) \]

(9)
where $\mathbf{B}_i$ and $\mathbf{h}$ are vectors of appropriate single-site operators and boundary fields. Since the system is not translation invariant, the one-point function $\langle A_n \rangle$ depends on the position $n$ near the chain ends, but approaches its bulk value in the middle of the system. If the system size is much larger than the correlation length the one-point function rapidly saturates and forms a plateau. Alternatively, if the chain length is too small compared to the correlation length (e.g., the system is critical or it is close to criticality) no plateau appears to develop.

There are two ways to estimate the order parameter and the associated correlation length from one-point functions. One can define the value $a_{L/2} \equiv \langle A_{L/2} \rangle$ measured in the middle of the chain and study its dependence on the total chain length $L$. It is expected that $a_{L/2}(L) \to a$ (with $a$ the bulk value of the order parameter) exponentially fast as $L \to \infty$, whenever the system is away from criticality, while the convergence is only algebraic at criticality. A disadvantage of this method is that many independent DMRG runs must be done with different $L$ values, or one is forced to use the “infinite lattice” algorithm which is less precise.

Alternatively, one can analyze the profiles $a_n \equiv \langle A_n \rangle$ as a function of $n$ at a fixed length $L$, providing that $L$ exceeds the correlation length $\xi$ reasonably (which can be checked a posteriori). In this latter case the profile is expected to have the following asymptotic form far from the chain ends

$$a_n = a + c e^{-n/\xi} + c e^{-(L+1-n)/\xi},$$

where $a$ is the bulk value of the order parameter, $\xi$ is the associated correlation length, $\chi$ is a suitable exponent, and $c$ is a constant. In general one can assume that the order parameter and the correlation length measured this way are identical to their bulk values derived from two-point correlation functions. However, the exponent of the polynomial prefactor $\chi$ is normally different from its bulk value and may depend on the boundary conditions chosen.

In practice we analyze our numerical data by dividing the chain into smaller segments with a length of the order of $\xi$, and perform local least-squares fits using the formula in Eq. (10). We have found it appropriate to fix $\chi$ first and then look for its value which makes the other fitting parameters the most stable in different segments as $n \to \infty$. Our procedure of determining order parameters and correlation lengths was extensively tested on a simpler problem, the 1D Ising model in a transverse field (ITF), where exact answers to many questions of interest are available. We summarize our experience with this model in the Appendix.

In most cases the DMRG is very efficient to calculate spectral gaps. The method is less useful, however, if the ground state is (asymptotically) highly degenerate, since then several states have to be targeted together, resulting in a rapid drop in accuracy. In the cubic model under investigation we expect a 6-fold ground-state degeneracy in the magnetic phase, and a 3-fold degeneracy in the quadrupolar phase, in accordance with the number of easy-axis directions for the two order parameters. The 6-fold degenerate case already exceeded our computational limitations, thus some of our conclusions with respect to that case are based on short chain exact diagonalization results only.

Errors in the quantitative predictions we report in the next sections stem from two sources, either from the limited accuracy of the numerical technique we have been using, or from the approximate (asymptotic) nature of the formulas we applied for fitting and extrapolating the data. As for the DMRG errors, our results are somewhat more precise when the “finite lattice” algorithm was used. In this case several iterations were done until convergence was reached. In principle, results obtained through the “infinite lattice” method inevitably contain a systematic “environment” error, and thus should be treated with less confidence. However, in the present problem improvements due to the “finite lattice” iterations turned out to be rather small, providing enough ground to assume that even our “infinite lattice” results have sufficient accuracy.

In general we made various runs keeping different number of states $M$, and extrapolated our results for $M \to \infty$, or at least checked that the results had been converged in this parameter. Typically, we found good convergence in $M$ despite the fact that the maximum number of states kept did not exceed $M = 85$. Note that the number of degrees of freedom at a single site is five in the $S = 2$ case, and the model in question has no continuous axial symmetry (total $S^z$ is not conserved) which could have been utilized to facilitate the DMRG calculation in the usual way.

IV. NUMERICAL RESULTS

In order to obtain the zero temperature phase diagram and critical properties of the model we carried out extensive numerical calculations using exact diagonalization techniques on short chains and the DMRG method on longer systems. In the case of open boundary conditions with boundary fields the actual form of the Hamiltonian used in the simulations was

$$H = - \sum_{i=1}^{L-1} \mathbf{S}_i \mathbf{S}_{i+1} - \sum_{i=1}^{L} D_i \left[ (S_i^z)^4 + (S_i^x)^4 + (S_i^y)^4 \right] - \mathbf{h} \cdot (\mathbf{B}_1 + \mathbf{B}_L),$$

with a reduced cubic crystal field at the chain ends (to counterbalance the missing bonds for the first and the last spins)

$$D_i = \begin{cases} D & \text{if } i = 2, \ldots, L-1 \\ D/2 & \text{if } i = 1, L \end{cases}$$

and a general symmetry-breaking boundary field containing magnetic and quadrupolar terms.
In order to avoid handling complex numbers, we did not include a boundary term proportional to $S_y$.

After the ground state had been found, we measured the order parameters. To facilitate further discussion we introduce the notations

$$m_n^\alpha = \langle S_n^\alpha \rangle, \quad q_n^\alpha = \langle (S_n^\alpha)^2 - 2 \rangle$$

with $n = 1, \ldots, L; \alpha = x, y, z$.

A. Ground state degeneracies and gaps

Figure 1(a) shows the lowest energy levels in a periodic chain with $L = 8$ sites obtained using exact diagonalization techniques. Although the chain is short, the 6-fold degeneracy, consisted of a singlet, a doublet, and a triplet is clearly discernible for small values of $D$. In the large $D$ (quadrupolar) region the degeneracy seems to be 3-fold, consisted of the singlet and the doublet only.

In both regimes we observe signs of a gap above these states, although the number of data points ($L = 4, 6, 8$) available for a given $D$ did not allow us to perform a detailed finite-size scaling study. Note that since the broken symmetry is discrete we do not expect any massless Goldstone modes in the ordered regimes. Excited states are rather massive domain walls, which separate domains with different ordering directions, and propagate in the system.

As was discussed in the former section, we could only apply the DMRG for the 3-fold degenerate case. We calculated the low-lying four states in an open chain at $D = 1.3$, which is above the critical point $D_m$ (see later sections), by targeting four states without any boundary fields. The results, presented in Fig. 1(b), strongly support the 3-fold asymptotic degeneracy with a finite gap $\Delta \approx 0.08 \pm 0.02$ above it in the thermodynamic limit. Unfortunately, this calculation was rather time consuming, which impeded us to repeat it for other $D$ values.

B. Magnetic order

Let us now investigate the spontaneous magnetization and the corresponding correlation length. In these calculations, purely magnetic boundary fields were used, i.e., $h_{xx} = h_{yy} = h_{zz} = 0$ in Eq. (13).

![Fig. 2](image-url)  

**FIG. 2.** (a) The parameter $m_{L/2}^x$ vs the chain length $L$, and (b) the profile $m_n^x$ vs the position $n$ at $L$ fixed, for some representative values of $D$. Purely magnetic boundary fields applied along the $x$ axis as given by $h_x$.

Expecting the magnetic easy axes along the cube edges we started with applying a boundary field along the $x$ axis setting $h_x = 10$ or 0.1, $h_z = 0$. Due to the symmetry, with such a boundary field one must obtain $m_n^x \neq 0$ and $m_n^y = m_n^z = 0$. Figure 2(a) shows how the parameter $m_{L/2}^x$, measured in the center of the chain, changes with the total chain length $L$ for representative values of the crystal-field strength $D$. One can see that for $D = 0.5, 1, 1.2$ the parameter $m_{L/2}^x$ converges to finite values independent of $h_x$. This clearly indicates a finite magnetic order for these $D$ values. On the other hand, for $D = 1.3, 1.5, 2$ the parameter $m_{L/2}^x$ goes to zero as $L \to \infty$, meaning no magnetic order. As characteristic examples the magnetization profiles $m_n^x$ with $L$ fixed are shown in Fig. 2(b) for $D = 1$ and 2. For $D = 1$ a plateau corresponding to a finite spontaneous magnetization $m^x \approx 1.76$, while for $D = 2$ a plateau with $m^x = 0$ can be observed.

So far we have assumed that the cube edges give the
easy axes in the magnetic phase. To verify this assumption, we also applied boundary fields which do not coincide with the expected easy directions. Choosing $h_x = 0.04$ and $h_z = 0.03$ we found that for $L \to \infty$ the parameter $m_{L/2}^\alpha$ always tends to the same value as found above, while $m_{L/2}^\gamma$ goes to zero, in agreement with Eq. (2).

![Graph](image)

**FIG. 3.** The spontaneous magnetization $m$ (left axis, dots) and the corresponding correlation length $\xi_m$ (right axis, triangles) as functions of $D$. Notice the large error bars for $\xi_m$ in the magnetic phase.

The above calculations allowed us to determine how the bulk spontaneous magnetization $m$ changes with the crystal-field strength $D$. We assume that monotonic dependencies of $m_{L/2}^\alpha$ on $L$, like those in Fig. 3(a), provide correct bounds for $m$, as was found for the ITF chain in the Appendix. The magnetization $m$ is plotted in Fig. 3 as a function of $D$, together with the corresponding correlation length $\xi_m$, discussed in details below. Since $\xi_m$ increases around the phase transition, very long chains have to be studied in order to find the large-$L$ limit of $m_{L/2}^\alpha$ in this region. The largest $D$ for which we can see the magnetic order is $D = 1.23$. For this $D$, taking $L = 400$, we obtain a finite $m$ with an uncertainty of 0.003 (for smaller $D$, shorter chains are sufficient to calculate $m$ much more precisely). Due to the increasing correlation length, and thus computational limitations, we were not able to trace the dependence of $m$ on $D$ further. Starting from $D = 1.26$, the phase with no magnetic order is observed.

Although it cannot be shown directly that the spontaneous magnetization disappears continuously at the phase-transition point, the sharp increase of the correlation length suggests that the transition is continuous. We therefore attempt to fit the dependence of $m$ on $D$, shown in Fig. 3, assuming a power law singularity and a linear regular term

$$m(D) = c_1(D_m - D)^\beta + c_2(D_m - D).$$

The fit is very good and we obtain $D_m = 1.2374 \pm 0.0004$, $\beta = 0.127 \pm 0.004$, $c_1 = 2.15 \pm 0.05$, $c_2 = -0.17 \pm 0.05$. Interestingly, the exponent $\beta$ is very close to that of the two-dimensional Ising model. The obtained parameter $D_m$ is our best estimate of the phase-transition point.

To calculate the magnetic correlation length $\xi_m$, we analyze the profiles of $m_{n}^\alpha$ at fixed $L$, analogous to those presented in Fig. 3(b). As was described earlier, we divide the chain into short sections and perform local fits using the formula in Eq. (4).

For each $D > D_m$, i.e., in the phase with no magnetic order, the local values of the fitting parameters obtained with $\chi = 0$ are very stable, and the convergence with increasing $n$ (towards the center of the chain) is convincing. The fitted order parameter $m$ is always very close to zero. It seems that the decay of $m_{n}^\alpha$ is asymptotically purely exponential, like the decay of $m_{n}^{\text{ITF}}$ in the disordered phase of the ITF chain, discussed in the Appendix. Due to the stability of local $\xi_m$, the correlation length $\xi_m$ in the phase with no magnetic order can be calculated very precisely (at $D = 1.27$ the error in $\xi_m$ is around 0.1% and it is smaller for larger $D$).

In the magnetically ordered phase, for $D < D_m$, the local fitting parameters obtained with $\chi = 0$ are not stable. The local $\xi_m$ increase towards the center of the chain, like in the ordered phase of the ITF chain, suggesting the existence of an algebraic prefactor. To calculate $\xi_m$, we vary $\chi$ and look for its value where the local $\xi_m$ are most stable. We do not find a universal prefactor, and the obtained $\chi$ depends on $D$ and $h_z$. However, the estimates of $\xi_m$ are independent of the boundary field, which justifies the procedure; the uncertainties in $\xi_m$ are around 10%. (The same procedure was tested for the ordered phase of the ITF chain and yielded acceptable estimates of the correlation length — see Appendix.)

The dependence of the correlation length $\xi_m$ on the crystal-field strength $D$ is plotted in Fig. 3. Due to the large error bars for $D < D_m$, the functional dependence can only be analyzed for $D > D_m$. Here, assuming a second order transition again, we fit the dependence of $\xi_m$ on $D$ with a power law

$$\xi_m(D) = c_3(D - D'_m)^{-\nu}.$$  

(16)

The fit is again rather good, yielding the values $D'_m = 1.236 \pm 0.004$, $\nu = 1.02 \pm 0.06$.

Thus, close to the phase transition both the spontaneous magnetization and the corresponding correlation length seem to be well described by power laws. The two independent estimates of the phase-transition point, $D_m$ and $D'_m$ from Eqs. (15) and (16), nicely coincide with each other. (In what follows, we will use $D_m$, which is more precise.) Moreover, the exponents $\beta$ and $\nu$ are close to those of the two-dimensional Ising model. Altogether, we have found strong indications that, as far as the magnetic quantities $m$ and $\xi_m$ are concerned, the phase transition is continuous and is described by the critical exponents of the two-dimensional Ising model.
C. Quadrupolar order

We now investigate the quadrupolar order, associated with the parameter $q^n$ defined in Eq. (3). While the quadrupolar order must be present in the magnetic phase ($D < D_m$), the question whether it exists in the phase with no magnetic order ($D > D_m$) is of central interest.

Expecting the quadrupolar easy axes along the cube edges, as given in Eqs. (6) and (8), we began with applying boundary fields along the $x$ axis. These were purely quadrupolar: $h_{nz} = 0; h_{xx} \neq 0, h_{yy} = h_{zz} = 0$ in Eq. (13); or purely magnetic: $h_{n} \neq 0, h_{zz} = 0, h_{xx} = 0$. With such boundary fields, due to the symmetry, one must obtain $q^n_x \neq q^n_z = q^n_n$.

![Figure 4](image)

**FIG. 4.** (a) The parameter $q_{L/2}^x$ vs the chain length $L$, and (b) the profile $q^n_x$ vs the position $n$ at $L$ fixed, for two values of $D$. Purely magnetic ($h_z$) and purely quadrupolar ($h_{xx}$) boundary fields applied along the $x$ axis.

We found that in both phases the order parameter $q^n_L$ measured in the chain center converges with increasing $L$ to finite values which do not depend on the actual boundary fields. Figure 4(a) shows these dependencies for $D = 1.2 < D_m$ and $D = 1.3 > D_m$. The appropriate profiles of $q^n_L$ with $L$ fixed are plotted in Fig. 4(b). In both phases the picture is typical for an ordered state: $q^n_L$ has plateaus in the middle of the chain, which correspond to finite bulk $q^n$. Thus, we find quadrupolar order on both sides of $D_m$, confirming the existence of a purely quadrupolar phase for $D > D_m$.

To verify that the cube edges give the easy axes of the quadrupolar ordering, we applied a quadrupolar boundary field having all three components different from zero: $h_{n} = 0, h_{xx} = 0.03, h_{yy} = 0.02$, and $h_{zz} = 0.01$. Then, in both phases, it was observed that $q_{L/2}^x > q_{L/2}^y = q_{L/2}^z$ in the large-$L$ limit, and the limiting values of $q_{L/2}^x$ were the same as found above, in accordance with Eqs. (6) and (8).

Assuming that monotonic dependencies of $q_{L/2}^y$ on $L$ provide correct bounds for the bulk value $q$, we determined $q$ for various values of $D$. ($q$ is defined as the largest of $q^n$ in a broken-symmetry state.) The results are depicted as bold points in Fig. 5, the precision in $q$ is $10^{-3}$ or better. We observed that, starting from $D \approx 1.3$, the associated correlation length $\xi_q$ increases rapidly with $D$ (see the inset in Fig. 5). At the same time, the DMRG precision for $q^n_L$ fell dramatically. For these reasons, already at $D = 1.45$ (and for larger $D$ too) we were unable to obtain precise results for a chain long enough to show us the large-$L$ limit of $q_{L/2}^n$ (for $D = 1.45$ and 1.5 we obtained upper bounds on $q$, which are shown as open circles in Fig. 5). We had similar problems close to the transition point $D_m$.

![Figure 5](image)

**FIG. 5.** The quadrupolar order parameter $q$ as a function of $D$. For $D = 1.45$ and $1.5$ upper bounds on $q$ are depicted as the open circles. The inset shows very rough estimates of the corresponding correlation length $\xi_q$.

The effective Hamiltonian $H_{eff}$ in Eq. (3) may help to understand the difficulties for $D > 1.4$. Notice, that the symmetry-breaking term $\tilde{J}_4$ in $H_{eff}$ is very small, $\tilde{J}_4/J_1 = O(1/D^2)$. Thus even if $\tilde{J}_4$ is relevant, the correlation length is expected to diverge rather rapidly as $D \rightarrow \infty$. On the other hand, the observed abrupt increase in $\xi_q$ and the emerging numerical difficulties beyond $D \sim 1.4$ may indicate an additional phase transition in which the quadrupolar long-range order vanishes, and gives rise to an extended critical phase with algebraically decaying quadrupolar correlations beyond a critical value $D_q$. A naive linear fit to the data points for $q$ in Fig. 5 would yield a value $D_q \approx 2.1$, but we are not in a position to give any firm claims in this question. Note however that if $D_q = \infty$, i.e., if the quadrupolar phase extends to $D \rightarrow \infty$, the curve $q(D)$ should finally bend upwards, and in the data points in Fig. 5, together with the upper bounds at $D = 1.45$ and 1.5, the (small) curvature is consistently downwards.

Now we analyze the dependence of $q$ on $D$ in Fig. 5 close to the phase-transition point $D_m$. For $D < D_m$ we assume a power-law singularity and a linear regular term

$$q(D) = q_{\text{left}} + c_4(D_m - D)\theta + c_5(D_m - D).$$

Fitting yields

$$q_{\text{left}} = 1.838 \pm 0.01, \qquad \theta = 0.5 \pm 0.1,$$
\[ c_4 = 0.44 \pm 0.1, \quad c_5 = -0.36 \pm 0.1. \] Thus, on this side of the phase-transition point, the quadrupolar order parameter appears to have singular behavior, but the exponent \( \theta \) differs from \( \beta \) describing the spontaneous magnetization.

For \( D > D_m \), we suppose the behavior is regular, which is suggested by Fig. 5(b). Our fitting formula is now

\[ q(D) = q_{\text{right}} + c_6(D - D_m) + c_7(D - D_m)^2, \]  

(18)

giving

\[ q_{\text{right}} = 1.840 \pm 0.002, \]
\[ c_6 = -1.83 \pm 0.03, \quad c_7 = -0.6 \pm 0.1. \] The left and right estimates of the order parameter \( q \) at \( D_m \) are in agreement, which supports our scaling assumptions. However, we cannot exclude the possibility that a singular derivative in \( q \) exists also for \( D > D_m \), so close to \( D_m \) that we do not observe it in our numerics.

To calculate the correlation length \( \xi_q \), we analyze the profiles of \( q_n^a \), like those in Fig. 5(b). In our opinion, only a purely quadrupolar boundary field is suitable for this purpose. With a magnetic boundary field, the decay of \( q_n^a \) could be driven by the decay of \( m_n^a \), and so governed by \( \xi_m \) and not \( \xi_q \).

Using the formula in Eq. 12 again, we find that both for \( D < D_m \) and \( D > D_m \) the prefactor exponent \( \chi \) is nonzero and it depends on \( D \) and the boundary field. The fitting yields rough estimates to \( \xi_q \) (Fig. 5 inset), the errors are believed to be around 20%. The results suggest that \( \xi_m = \xi_q \) for \( D < D_m \), which is the expected answer, since the magnetic and quadrupolar order is intimately connected in this phase. For \( D > D_m \), the correlation length \( \xi_q \) increases with \( D \) starting from \( D \approx 1.3 \), as mentioned above, but, due to the poor precision, we cannot resolve whether \( \xi_q \) diverges (numerically it increases sharply) for \( D \to D_m \) or it remains finite.

V. CONCLUSIONS

In summary, we have studied the ground-state properties of a one-dimensional \( S = 2 \) ferromagnetic spin chain with single-site cubic crystal-field anisotropy \( D \). We argued that in contrast with the mean-field prediction, perturbation theory suggests the possibility that a purely quadrupolar phase exists for large values of \( D \). This quadrupolar phase was expected to be separated from the magnetic phase by a continuous quantum phase transition at a critical point \( D_m \). We verified this conjecture by investigating the model numerically using the density-matrix renormalization group method on open chains with special, symmetry-breaking boundary conditions.

In most cases, the method provided precise estimates of the magnetic and quadrupolar order parameters. Very close to the phase-transition point and in the large-\( D \) limit, however, our results were less accurate due to the rapidly increasing correlation lengths. For the correlation lengths themselves we could only obtain rather rough estimates.

Evidence has been obtained that, in qualitative agreement with the mean-field prediction, the spontaneous magnetization diminishes continuously at the phase-transition point. Regarding the magnetic properties, the transition seems to be characterized by the critical exponents of the two-dimensional Ising model. This could be plausible, since the extra broken symmetry of the magnetic ground state with respect to that of the quadrupolar ground state is just a \( Z_2 \) subgroup — the same group as the one breaking down spontaneously in the case of the Ising model.

Both in the magnetic and nonmagnetic phases we demonstrated the presence of a quadrupolar order. In the former case the quadrupolar order is just a “secondary” effect, inevitably present in any \( S \geq 1 \) model with magnetic order. In the latter case, however, it is the “primary” order, constituting a qualitatively different broken-symmetry phase. At the phase-transition point \( D_m \), the quadrupolar order parameter is continuous. For \( D \lesssim D_m \), we could clearly discern a singularity in the derivative of the order parameter, and found that the quadrupolar correlation length diverges together with the magnetic correlation length. For \( D \gtrsim D_m \), our observations are much less concrete: the order parameter can be fitted reasonably well by a low-order (regular) polynomial without any singular terms, and, although the quadrupolar correlation length increases as \( D_m \) is approached from above, our results do not suggest unambiguously a divergence on this side. It is not clear for us whether the phase transition involving the magnetization should have any precursor in the quadrupolar fluctuations above \( D_m \).

Mainly due to computational limitations we were unable to resolve convincingly the question whether the quadrupolar phase extends to \( D \to \infty \) or there is a finite value \( D_q \) where the quadrupolar long-range order disappears. This question is unique to the one-dimensional situation — in two dimensions and higher we expect a finite quadrupolar order at zero temperature for any \( D > D_m \).

Finally, the scenario for the magnetic-to-quadrupolar phase transition was supported by the analysis of the degeneracy of the ground state as a function of \( D \). We observed a 6-fold and a 3-fold asymptotic degeneracy below and above \( D_m \) respectively, in accordance with the the number of possible ordering directions in the two phases.

The confirmation of the purely quadrupolar phase in the one-dimensional case gives rise to the belief that such a phase should also exist in higher dimensions at sufficiently low temperatures when the cubic crystal field is strong. Increasing temperature necessarily destroys the quadrupolar order and leads to a finite-temperature phase transition between the quadrupolar and a completely disordered phase. Note that the correct critical theory of this transition cannot be obtained by simply substituting spins with classical vectors in our model, since this approach is unable to account for the purely quadrupolar order. The usual practice of neglecting quantum fluctuations by treating spins classically around
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APPENDIX: A TEST CALCULATION

Our numerical procedure and extrapolation methods were tested on the zero-temperature 1D Ising model in a transverse field (ITF) where an exact solution for the bulk quantities is available. Our experience with this model provided useful guidelines to refine our procedure in the study of the cubic model of Eq. (1).

The Hamiltonian of the ITF model on an open chain with length $L$ is defined as

$$H_{\text{ITF}} = -\sum_{i=1}^{L-1} \sigma_i^x \sigma_{i+1}^x - \sum_{i=1}^{L} \Gamma_i \sigma_i^x$$

(A1)

$$\Gamma_i = \begin{cases} \Gamma & \text{if } i = 2, \ldots, L - 1 \\ \Gamma/2 & \text{if } i = 1, L \end{cases}$$

where $\sigma_i^\alpha$ denotes Pauli matrices at site $i$, and $\Gamma$ is the transverse magnetic field. Note that in the above definition the spins at the chain ends, being coupled to one neighbor only, are subject to the reduced field $\Gamma/2$. We concentrate on the spontaneous magnetization $m_{\text{ITF}} = \langle \sigma_z \rangle$ and the corresponding correlation length $\xi_{\text{ITF}}$. For $\Gamma = 0$ the Hamiltonian $H_{\text{ITF}}$ has the classical, ferromagnetic ground state with $m_{\text{ITF}} = \pm 1$, and the single-site term in $H_{\text{ITF}}$ tends to destroy the spontaneous magnetization; in this respect the ITF model is similar to the cubic model in Eq. (1). In the thermodynamic limit there are two phases in the ITF chain, distinguished by the order parameter $m_{\text{ITF}}$. For $0 < \Gamma < 1$ the ground state is ordered due to spontaneous symmetry breaking

$$m_{\text{ITF}} = \pm \left(1 - \Gamma^2\right)^{1/8}, \quad \xi_{\text{ITF}} = -\frac{1}{2 \log \Gamma}$$

(A2)

while for $\Gamma > 1$ there is a disordered phase with

$$m_{\text{ITF}} = 0, \quad \xi_{\text{ITF}} = \frac{1}{\log \Gamma}$$

(A3)

At $\Gamma = 1$ the ground state is critical and $\xi_{\text{ITF}} = \infty$.

In our study the following values of the transverse field were considered: $\Gamma = 0.9$ (ordered phase) where $\xi_{\text{ITF}} \approx 4.75$; $\Gamma = 1.1$ (disordered phase) where $\xi_{\text{ITF}} \approx 10.49$; and the critical point $\Gamma = 1$ with $\xi_{\text{ITF}} = \infty$. We computed the position dependent order parameter $m_{\text{ITF}} = \langle \sigma_z \rangle$ using the DMRG method with boundary fields up to $L = 200$. As a symmetry-breaking boundary field we considered

$$B = \sigma_z$$

(A4)

in Eq. (1); and we used two different values of $h$: a strong $h = 10$ and a weak $h = 0.1$ boundary field. In the case of the ITF chain the DMRG is extremely precise even close to the critical point. Keeping a relatively small number of states $M = 64$ the truncation errors are already negligible and the fitting procedure can be tested on numerically exact data. We emphasize that although rigorous results are available for quantities in the thermodynamic limit, the behavior of relatively short chains with complicated boundary conditions is not feasible for a study with purely analytical tools even for the ITF model.

The value of the order parameter at one of the central spins $m_{L/2}$ as a function of the chain length $L$ is shown in Fig. 6(a). For these data the “infinite-lattice” DMRG was used from $L = 4$ to $L = 200$, thus a systematic “environment error” cannot be excluded. In the ordered phase, $m_{L/2}$ converges to the same finite value for both values of $h$, and the two dependencies are monotonic. At each $L$, correct bounds for the bulk $m_{\text{ITF}}$ in Eq. (A2) are obtained, and at $L = 200$, ten digits of $m_{\text{ITF}}$ are recovered. In the disordered phase, $m_{L/2}$ converges to zero for both boundary fields $h$. At the critical point, the limit of $m_{L/2}$ does not show up in Fig. 6(a), we can only say that the correlation length must be of the order of $L = 200$ or larger.

In Fig. 6(b) we show the order parameter profiles $m_n$ for a chain length fixed at $L = 200$. This length exceeds the exact correlation lengths $\xi_{\text{ITF}}$ for $\Gamma = 0.9$ and $\Gamma = 1.1$ many times. There are plateaus for $\Gamma = 0.9$ and
\[ \Gamma = 1.1, \] which correspond to the bulk values of the order parameter. There is no plateau at the critical point where the correlation length is infinite and the dependence of the profile is algebraic.

In order to calculate the correlation length \( \xi \), we analyze the profiles from Fig. 3(b) using the ansatz in Eq. (10). As is described in Sec. III we first fix a value of the exponent \( \chi \) and then fit for the other parameters. Repeating the fits in different sections of the chain we seek the value of \( \chi \) which makes the other parameters the most stable as a function of \( n \).

For \( \Gamma = 1.1 \) the value of the exponent which yields the most stable fitting parameters is \( \chi \approx 0 \). From positions \( n = 50 \) to \( n = 100 \), and for both \( h \), the fitted correlation length \( \xi \) agrees with the exact \( \xi \) with a precision of 7 digits! It seems that, asymptotically, the decay of \( m_{\text{ITF}} \) in the disordered phase is purely exponential. This should be compared with the exponent \( \chi_{\text{bulk}} = 1/2 \) characterizing the decay of the bulk two-point correlation function in this phase. (For a similar difference in algebraic prefactors of one and two-point correlation functions in an \( S = 1 \) chain see Ref. 13.)

In the ordered phase for \( \Gamma = 0 \) we can also start with \( \chi = 0 \). In this case, however, the fitting parameters are not stable, and \( \xi \) increases with \( n \) for both values of \( h \), as is illustrated by Fig. 4. This suggests that a nonzero prefactor is present. The value of \( \chi \) which makes the correlation length \( \xi \) the most stable seems to depend on the boundary field applied. We obtain \( \chi \approx 1.25 \pm 0.05 \) for \( h = 10 \) and \( \chi \approx 0.9 \pm 0.05 \) for \( h = 0.1 \), we cannot see a universal prefactor. (Note that for the bulk two-point functions the exponent of the prefactor is \( \chi_{\text{bulk}} = 2 \) in this phase.) The values of \( \xi \) calculated with the above prefactors are shown in Fig. 3. It is seen that using these prefactors we find better estimates for \( \xi \) than with \( \chi = 0 \).

Anticipating computational limitations in the investigation of the \( S = 2 \) cubic model in Eq. (11), we also analyzed shorter chains, such that \( L \approx 10^2 \) during the test calculations. For \( \Gamma = 1.1 \) and \( L = 100 \) four digits of the exact \( \xi \) are recovered; for \( \Gamma = 0.9 \) and \( L = 50 \) only round estimates for \( \xi \) are obtained, the error is around 10%.

Our observations can be summarized as follows:

- The order parameter \( m_{\text{ITF}} \) can be calculated accurately in both phases; it can be decided whether the system is ordered or not.
- In the ordered phase, upper and lower bounds for \( m_{\text{ITF}} \) are obtained by considering strong and weak boundary fields, respectively.
- The correlation length \( \xi \) can be calculated very precisely in the disordered phase.
- In the ordered phase, rather rough estimates of \( \xi \) can be found.

**FIG. 7.** ITF chain results. Ordered phase \( \Gamma = 0.9 \). The local values of the correlation length \( \xi \) from Eq. (10), obtained with different boundary fields \( h \) and exponents \( \chi \), as a function of the position \( n \) at \( L = 200 \). The exact correlation length \( \xi_{\text{ITF}} \) is shown as a horizontal line.

On leave from the Research Institute for Solid State Physics, Budapest, Hungary.

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