Spin-distribution functionals and correlation energy of the Heisenberg model

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We analyze the ground-state energy and correlation energy of the Heisenberg model as a function of spin, both in the ferromagnetic and the antiferromagnetic case, and in one, two and three dimensions. First, we present a comparative analysis of known expressions for the ground-state energy $E_0(S)$ of *homogeneous* Heisenberg models. In the one-dimensional antiferromagnetic case we propose an improved expression for $E_0(S)$, which takes into account Bethe-ansatz data for $S = 1/2$. Next, we consider *inhomogeneous* Heisenberg models (e.g., exposed to spatially varying external fields). We prove a Hohenberg-Kohn-like theorem stating that in this case the ground-state energy is a functional of the spin distribution and that this distribution encapsulates the entire physics of the system, regardless of the external fields. Building on this theorem, we then propose a local-density-type approximation that allows us to utilize the results obtained for homogeneous systems in inhomogeneous situations too. We conjecture a scaling law for the dependence of the correlation functional on dimensionality, which is well satisfied by existing numerical data. Finally, we investigate the importance of the spin-correlation energy by comparing results obtained with the proposed correlation functional to ones from an uncorrelated mean-field calculation, taking as our example a linear spin-density wave state.

I. INTRODUCTION

In this paper we study the ground-state energy, correlation energy, and related quantities of the Heisenberg model. The homogeneous Heisenberg model is defined by the Hamiltonian

$$\hat{H}_0 = J \sum_{\langle ij \rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j , \qquad (1)$$

where $\hat{\mathbf{S}}_i$ is a spin vector operator satisfying $\hat{\mathbf{S}}_i^2 |Sm\rangle = S(S+1)|Sm\rangle$ and $\hat{S}_{i,z}|Sm\rangle = m|Sm\rangle$, and S and m are the spin quantum numbers of the particles under study. Although in accordance with common terminology the, $\hat{\mathbf{S}}_i$'s are called spin operators, they really represent total angular momentum and are not restricted to be of purely spin origin. $\langle ij \rangle$ indicates a sum over nearest neighbors on a lattice of dimensionality d and J is the spin-spin interaction constant, parametrizing the exchange interaction of the underlying microscopic Hamiltonian.[1-3] For antiferromagnetism $J > 0$, while for ferromagnetism $J < 0$. This model was originally proposed in 1926 to explain ferromagnetism in transition metals,[4,5] but has since found a large number of other applications to the magnetic properties of matter.[1-3] Recent examples are antiferromagnetic chains in complex oxides and other low-dimensional magnets[6] or studies of magnetic effects on crystal-field splittings in rare-earth compounds.[7]

The inhomogeneous Heisenberg model, characterized by broken translational symmetry, is obtained by adding a spatially varying magnetic field to $\hat{H}_0$,

$$\hat{H} = J \sum_{\langle ij \rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j + \sum_i \mathbf{B}_i \cdot \hat{\mathbf{S}}_i , \qquad (2)$$

where $\mathbf{B}_i$ can either be an externally applied field or an internal field due to magnetism in the system. A variety of different sources and manifestations of the inhomogeneity $\mathbf{B}_i$ has been studied in the recent literature, often in conjunction with the synthesis and investigation of real materials, whose magnetic properties are necessarily spatially inhomogeneous, but still to some extent describable by modified Heisenberg models.[6,8] The interpretation of experimental data for realistic materials in terms of the Heisenberg model, in particular in the presence of staggered or otherwise spatially varying magnetic fields, must be based on a solid understanding of the behavior of the model (2) in the presence of inhomogeneity.

The homogeneous Heisenberg model in $d = 1$ dimension and for $S = 1/2$ has an exact analytical solution in terms of the Bethe ansatz,[9,10] but the same ansatz does not work in higher dimensions, in which no exact solution is known. It is also hard to generalize to inhomogeneous situations. In the present paper we combine exact and approximate results obtained within a variety of different approaches and techniques, to provide a systematic analysis of the ground-state and correlation energy of the Heisenberg model in both the ferromagnetic and antiferromagnetic cases and for $d = 1, 2, 3$ dimensions.

In Sec. II we provide a comparative analysis of available expressions for the ground-state energy of the homogeneous Heisenberg model as a function of spin S, dimenionality d, and coupling constant J. This section also contains a proposal for an improved expression that goes beyond those available in the literature in taking into account a Bethe ansatz result for $S = 1/2$. In Sec. III we use concepts of density-functional theory to extend the utility of the homogeneous

results discussed in the preceeding section to inhomogeneous situations. This section includes a proof of a Hohenberg-Kohn-type theorem for a large class of generalized Heisenberg models, and a proposal for a simple local-density approximation. In Sec. IV we then study the correlation energy in homogeneous and inhomogeneous Heisenberg models, in order to assess the importance of correlations and quantum fluctuations as a function of dimensionality and spin. As an explicit example for a physically interesting type of inhomogeneity we consider a linear spin-density wave, and explore the differences between a mean-field and a density-functional treatment of the resulting inhomogeneous spin distribution.

II. HOMOGENEOUS HEISENBERG MODELS: GROUND-STATE ENERGY AS FUNCTION OF SPIN

This section provides a brief review of what is known about the ground-state energy of homogeneous Heisenberg models. Although the expressions collected below for $J > 0$ and $J < 0$ and $d = 1, 2, 3$ are given in various places in the literature, we have not found a systematic collection and comparison in one single place. For the convenience of the reader and future reference we therefore provide such a comparison below. We also add an expression to the list [Eq. (15)], which is a slight improvement on one of the earlier results.

A. Ferromagnetic case

Let us first consider the ferromagnetic case, which is much simpler than the antiferromagnetic one. At zero temperature all spins are parallel and the corresponding spin operators commute with each other. The exact ground-state energy is then the same as that obtained in the mean-field approximation and is given by

$$E_0^{FM}(S, J, z) = J \frac{z}{2} N S^2, \quad (3)$$

where z is the number of nearest neighbors, which on linear, square, and cubic lattices is related to the dimensionality by $d = z/2$ and, as above, $J < 0$ for ferromagnetism. In Fig. 1 we show the resulting curves for the energy per site and interaction strength

$$e_0^{FM}(S, d) = \frac{1}{N|J|} E_0^{FM}(S, J, z = 2d) = -dS^2, \quad (4)$$

for one, two, and three dimensions.

B. Antiferromagnetic case

The antiferromagnetic (AFM) case is much more complicated than the ferromagnetic one, because in spite of its name the ground-state of the antiferromagnetic Heisenberg model is not simply the "antiferromagnetic" state consisting of alternating spin up and spin down states with respect to a fixed direction (i.e., the Néel state), but a quantum superposition of states involving also spins along the perpendicular axes. In the one-dimensional $S = 1/2$ case the structure of the
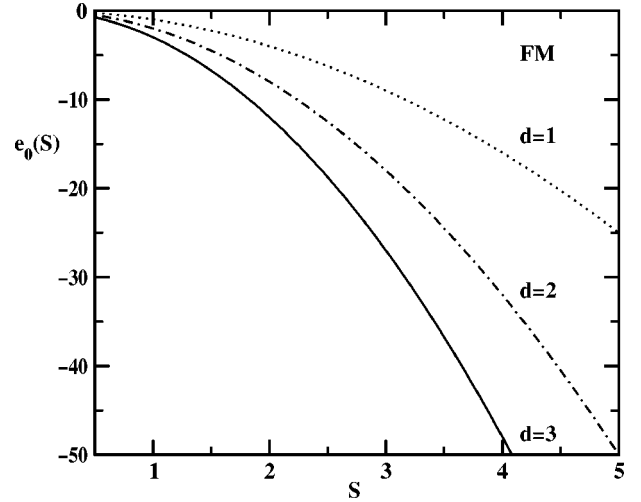

FIG. 1. Per-site ground-state energy of the homogeneous ferromagnetic Heisenberg model in one, two, and three dimensions.

corresponding ground-state for $N \to \infty$ is known exactly by means of the Bethe ansatz.[3,9,10] The corresponding ground-state energy is

$$E_0^{AFM}\left(S = \frac{1}{2}, J, d = 1\right) = E_0^{FM}\left(S = \frac{1}{2}, J, z = 2\right) - JN\ln 2. \quad (5)$$

For the energy per site and interaction strength one obtains from this[10]

$$e_0^{AFM}\left(S = \frac{1}{2}, d = 1\right) = \frac{1}{NJ} E_0^{AFM}\left(S = \frac{1}{2}, J, d = 1\right) \quad (6)$$

$$= \frac{1}{4} - \ln 2 = -0.443\,147\,18. \quad (7)$$

Similar exact results are not available in the general case, for arbitrary S and d. However, a set of useful approximate expressions was derived in two early papers by Anderson.[11,12] In the first of these it is shown by means of a variational argument that the energy of the antiferromagnetic ground state must lie in the interval[11]

$$-\frac{z}{2} S^2 \left(1 + \frac{1}{zS}\right) \leq e_0^{AFM}(S, z) \leq -\frac{z}{2} S^2, \quad (8)$$

where $z = 2d$ is again the coordination number of the linear, square or cubic lattice under study. A simple estimate is obtained by using the center of this interval,[12] i.e.,

$$e_0^{AFM}(S, z) \approx -\frac{z}{2} S^2 \left(1 + \frac{1}{2zS}\right), \quad (9)$$

but the quality of this estimate, which is quite good for $d = 3$, deteriorates for $d = 2$ and $d = 1$. A numerical calculation based on spin-wave theory leads to the more precise results[12]

$$e_0^{\text{AFM}}(S, d=1) = -S^2 + \left(\frac{2}{\pi} - 1\right)S \qquad (10)$$

$$= -S^2\left(1 + \frac{0.363\,38}{S}\right), \qquad (11)$$

$$e_0^{\text{AFM}}(S, d=2) = -2S^2\left(1 + \frac{0.158}{S}\right), \qquad (12)$$

$$e_0^{\text{AFM}}(S, d=3) = -3S^2\left(1 + \frac{0.097}{S}\right). \qquad (13)$$

We can assess the quality of these expressions by substituting $S = 1/2$ in the first of them. The result $e_0^{\text{AFM}}(S = \frac{1}{2}, d = 1) = -0.431\,69$ is within 2.6% of the exact Bethe ansatz value reported above.

Indeed, significant improvement over Eq. (10) has only been obtained recently, with the use of modern computing facilities and advanced numerical techniques. In such work Lou *et al.*[13] used (50 years after Ref. 12) the density-matrix renormalization group (DMRG) to calculate corrections to Eq. (10) for values of S ranging from 1/2 to 5 in steps of 1/2. These authors propose a fit to their numerical data which in our present notation reads

$$e_0^{\text{AFM,fit1}}(S, d=1) = -S^2 + \left(\frac{2}{\pi} - 1\right)S - 0.03262 - 0.0030\frac{1}{S}$$

$$- \left(0.338 - \frac{0.28}{S}\right)e^{-\pi S}\cos(2\pi S). \qquad (14)$$

We note that for $S = 1/2$, where it predicts $e_0^{\text{AFM,fit1}}(S = \frac{1}{2}, d = 1) = -0.516\,459$, this fit is actually worse than the earlier expression (11), deviating by 17% from the exact Bethe ansatz value. On the other hand, as shown in Ref. 13 by comparison with DMRG data and other highly precise numerical results, the fit is excellent for higher values of S.

In order to obtain a closed expression that can also be applied at $S = 1/2$, a slight modification to the fit by Lou *et al.* is sufficient. To this end we propose the alternative expression

$$e_0^{\text{AFM,fit2}}(S, d=1)$$

$$= -S^2 + \left(\frac{2}{\pi} - 1\right)S - 0.032\,62 - 0.0030\frac{1}{S} + 0.0015\frac{1}{S^3}$$

$$- \left(0.338 - \frac{0.28}{S} + \frac{0.035}{S^3}\right)e^{-\pi S}\cos(2\pi S), \qquad (15)$$

which differs from Eq. (14) by the inclusion of two cubic terms in $1/S$. The value at $S = 1/2$ predicted by this expression $e_0^{\text{AFM,fit2}}(S = \frac{1}{2}, d = 1) = -0.446\,253$, deviates by only 0.7% from the exact Bethe ansatz value.

Figures 2 and 3 display the various AFM energy expressions collected above. In Fig. 2 we compare the rigorous but rather wide interval provided by expression (8), with the
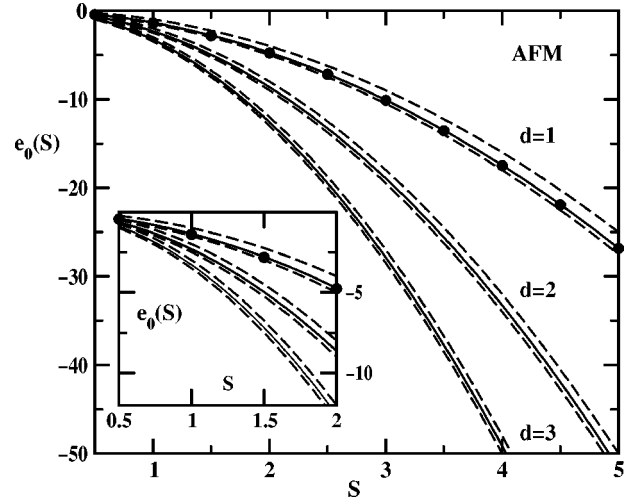

FIG. 2. Per-site ground-state energy of the homogeneous antiferromagnetic Heisenberg model in one, two, and three dimensions. Dashed lines: interval according to Eq. (8). Full lines: spin-wave theory expressions (11), (12), and (13), respectively. Full circles (in one dimension only): DMRG data of Ref. 13. The inset is a zoom into the low spin region of the main figure.

spin-wave results (11)–(13) and (in $d = 1$) the DMRG data of Ref. 13. Obviously, on this scale the spin-wave expression (11) already provides an excellent approximation to the DMRG data. Neither of the two fits (14) and (15) can be distinguished from the much simpler expression (11) on the scale of the figure. Unfortunately, no highly precise numerical reference data, similar to the DMRG results of Ref. 13, seem to be available in $d = 2$ and $d = 3$, but the approximations leading to the simple analytical formulas given above are expected to work better as d increases. This expectation is corroborated by noting that the rigorous interval (8) shrinks with increasing d.

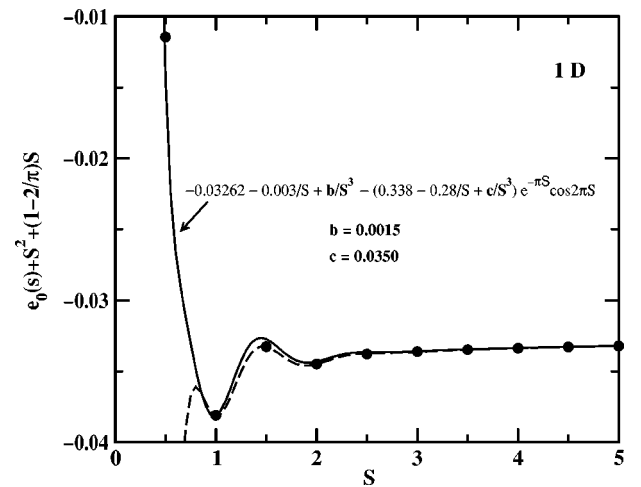Interestingly, both the numerically highly precise DMRG

FIG. 3. Same as in Fig. 2, but on a reduced scale. On the vertical axis the expression (11) has been subtracted to display only the corrections to that result. Full circles: DMRG data of Ref. 13, dashed line: fit proposed in that reference, present Eq. (14), full line: fit proposed here, Eq. (15).

data of Ref. 13 (in $d=1$) and the spin-wave expressions (11)–(13) (in $d=1,2,3$) systematically lie closer to the more negative boundary of the interval than to the less negative one, and in $d=1$ the DMRG values are still a little closer to this boundary than the curve predicted by Eq. (11). This shows that the lower bound in Eq. (8) is tighter than the upper one. The simple estimate (9), on the other hand, by construction falls in the middle of the interval and becomes less reliable for lower d. In the interest of readability we have not displayed the curves corresponding to this estimate in the figures.

Figure 3 shows that on a smaller scale the differences between the more precise expressions become important. Here we compare the two fits (14) and (15) to the DMRG data. To make the details of the fits, and the interesting oscillatory structure they display, clearly visible, we have subtracted the spin-wave expression (11), which is common to both fits. The fit proposed above, Eq. (15), is slightly inferior to the one developed by Lou *et al.*, Eq. (14), around $S = 3/2$, but unlike the latter recovers the exactly known $S = 1/2$ data point to within less than one percent.

In the present paper we are mainly concerned with the homogeneous or inhomogeneous Heisenberg model on linear, square, and cubic lattices. Expressions (or numerical values) for the ground-state energy can also be derived for many other variations of the Heisenberg model, such as lattices with helical boundary conditions,[14] or with anisotropic interactions.[15] Although we do not consider such models in the present paper, many of our results can be extended to them in a straightforward way.

III. INHOMOGENEOUS HEISENBERG MODELS: SPIN-DISTRIBUTION FUNCTIONALS

Based on the analysis of the preceeding section we recommend the use of expressions (11)–(13) in calculations requiring simple expressions for the ground-state energy of the homogeneous antiferromagnetic Heisenberg model in one, two, and three dimensions. In one dimension, where the simple expressions fare worst, either of the two fits (14) and (15) provides a significant improvement in accuracy, but only the latter recovers the Bethe ansatz value at $S=1/2$. However, the utility of any of these expressions is rather limited due to the restriction to spatial homogeneity. Externally applied magnetic fields that vary in space, calculations of magnetic effects on crystal-field splitting, description of nontrivial internal order, etc., require use of the *inhomogeneous* Heisenberg model.[8] Unfortunately, if translational invariance is broken the Bethe ansatz, spin-wave theory, DMRG, and most other approaches encounter very significant computational difficulties.

In the case of *ab initio* calculations a many-body technique that has had considerable success in the application to inhomogeneous systems is density-functional theory (DFT),[16–18] but it is not very common to apply DFT to model Hamiltonians also. However, following pioneering work by Gunnarsson and Schönhammer,[19] DFT was recently formulated and applied for the one-dimensional Hubbard model.[20–22] In this section we build on this experience to

explore how DFT can become a useful tool also in studies of the inhomogeneous Heisenberg model. To this end we prove, in Sec. III A, a Hohenberg-Kohn-type theorem for a wide class of generalized Heisenberg models (of which the models discussed above are special cases). In Sec. III B we then use this theorem and the explicit expressions discussed in Sec. II to construct a simple local-density approximation for inhomogeneous Heisenberg models.

A. Hohenberg-Kohn theorem for generalized Heisenberg models

A first question that must be answered before DFT can be usefully employed is what the fundamental variable is. In *ab initio* DFT one mostly chooses the particle density $n(\mathbf{r})$ or its spin-resolved counterpart $n_\sigma(\mathbf{r})$,[16–18] although other choices are occasionally useful.[23–25] In the case of the Hubbard model the basic variable is the site occupation number n_i.[19–22] In the present case we propose to use the spin vector $\mathbf{S}_i$, which is the only fundamental dynamical variable appearing in the definition of the Heisenberg model.

In the interest of generality, in the present section we consider a generalized Heisenberg model of the form

$$\hat{H}_g = \sum_{ij} J_{ij}\hat{\mathbf{S}}_i\cdot\hat{\mathbf{S}}_j + \sum_i \hat{\mathbf{S}}_i\cdot\mathbf{B}_i . \qquad (16)$$

Unlike in Eqs. (1) and (2) the sum in the first term on the right-hand side is not restricted to nearest neighbors, and the interaction J_{ij} can depend in any way on the indices of the involved sites. In particular, it can extend to next-nearest neighbors and beyond, or alternate between ferromagnetic and antiferromagnetic along some direction in the crystal. Both of these features are found in realistic magnetic crystals. This relaxation of constraints on J may appear a considerable complication, but it turns out that the proof of the Hohenberg-Kohn theorem is essentially unaffected by the extra generality. (As pointed out above, we do not consider anisotropic Heisenberg models, in which J couples differently to different components of $\mathbf{S}$, in this paper, but the generalization of the theorem to this case is straightforward.)

Following the steps of Hohenberg and Kohn, we now consider two Hamiltonians with same interaction J_{ij}, but exposed to two different magnetic fields $\mathbf{B}_i$ and $\mathbf{B}_i'$. Thus

$$\hat{H}\Psi = \left[\sum_{i,j} J_{ij}\hat{\mathbf{S}}_i\cdot\hat{\mathbf{S}}_j + \sum_i \hat{\mathbf{S}}_i\cdot\mathbf{B}_i\right]\Psi = E_0\Psi, \qquad (17)$$

$$\hat{H}'\Psi' = \left[\sum_{i,j} J_{ij}\hat{\mathbf{S}}_i\cdot\hat{\mathbf{S}}_j + \sum_i \hat{\mathbf{S}}_i\cdot\mathbf{B}_i'\right]\Psi' = E_0'\Psi', \qquad (18)$$

where E_0 and E_0' are the ground-state energies in the fields $\mathbf{B}_i$ and $\mathbf{B}_i'$, and Ψ and Ψ' are the corresponding ground-state wave functions.

As a consequence of the variational principle we have the inequality

$$E_0 = \langle\Psi|\hat{H}|\Psi\rangle < \langle\Psi'|\hat{H}|\Psi'\rangle, \qquad (19)$$

since Ψ' is not the ground-state wave function belonging to $\hat{H}$ (assumed nondegenerate). By adding and subtracting the term $\Sigma_i \hat{\mathbf{S}}_i \cdot \mathbf{B}_i'$ on the right-hand side, the inequality becomes

$$E_0 = \langle \Psi | \hat{H} | \Psi \rangle < \langle \Psi' | \hat{H}' | \Psi' \rangle + \sum_i \langle \Psi' | \hat{\mathbf{S}}_i \cdot (\mathbf{B}_i - \mathbf{B}_i') | \Psi' \rangle. \tag{20}$$

Here the first term on the right-hand side is just the ground-state energy E_0', of Hamiltonian $\hat{H}'$. With the abbreviations $\Delta \mathbf{B}_i = \mathbf{B}_i - \mathbf{B}_i'$, $\mathbf{S}_i = \langle \Psi | \hat{\mathbf{S}}_i | \Psi \rangle$, and $\mathbf{S}_i' = \langle \Psi' | \hat{\mathbf{S}}_i | \Psi' \rangle$ the preceeding equation then becomes

$$E_0 < E_0' + \sum_i \mathbf{S}_i' \cdot \Delta \mathbf{B}_i. \tag{21}$$

Now we repeat the same argument starting with the Hamiltonian $\hat{H}'$. The variational principle guarantees that

$$E_0' = \langle \Psi' | \hat{H}' | \Psi' \rangle < \langle \Psi | \hat{H}' | \Psi \rangle. \tag{22}$$

By adding and subtracting the term $\Sigma_i \hat{\mathbf{S}}_i \cdot \mathbf{B}_i$, we obtain, in the same way as before,

$$E_0' < E_0 - \sum_i \mathbf{S}_i \cdot \Delta \mathbf{B}_i. \tag{23}$$

Addition of Eqs. (21) and (23) leads to

$$E_0 + E_0' < E_0 + E_0' + \sum_i (\mathbf{S}_i' - \mathbf{S}_i) \Delta \mathbf{B}_i. \tag{24}$$

If we now assume that $\mathbf{S}_i' = \mathbf{S}_i$, i.e., that the two spin distributions corresponding to the two different wave functions Ψ and Ψ' are identical, then the previous equation reduces to the contradiction

$$E_0 + E_0' < E_0 + E_0'. \tag{25}$$

This contradiction shows that two distinct nondegenerate ground states can never lead to the same spin distribution. Hence, given some arbitrary spin distribution $\mathbf{S}_i$ there is *at most one* wave function which gives rise to it. In other words: the spin distribution uniquely determines the wave function. This means that the wave function is a functional[26] of the spin distribution, i.e., $\Psi = \Psi[\mathbf{S}_i]$. This is the statement of the Hohenberg-Kohn theorem for the Heisenberg model.

For completeness we mention that the above proof by contradiction, patterned after the one first presented by Hohenberg and Kohn in the *ab initio* case,[27] is not the only possible one. The constrained-search technique of Levy[28] and Lieb[29] is also easily adapted to the present case. The ground-state wave function in this approach is uniquely defined by its spin distribution as the wave function that minimizes $\langle \Psi | \hat{H} | \Psi \rangle$ and reproduces $\mathbf{S}_i$. This minimization defines the functional

$$E_{LL}[\mathbf{S}_i] = \min_{\Psi \to \mathbf{S}_i} \langle \Psi | \hat{H} | \Psi \rangle, \tag{26}$$

whose minimum is the ground-state energy.

An immediate consequence of either formulation of the proof is that the ground-state expectation value of any observable $\hat{O}$, is also a functional of the spin distribution, defined via

$$O[\mathbf{S}_i] = \langle \Psi[\mathbf{S}_i] | \hat{O} | \Psi[\mathbf{S}_i] \rangle, \tag{27}$$

and this functional is the same regardless of the strength and direction of the magnetic field $\mathbf{B}_i$, i.e., it is universal with respect to external fields. Note that the theorem applies to any ground-state observable. For example, it implies that also all multispin correlation functions of the general form

$$C_{n,n+1,n+2, \ldots} := \langle \Psi | \hat{\mathbf{S}}_n \hat{\mathbf{S}}_{n+1} \hat{\mathbf{S}}_{n+2} \cdots | \Psi \rangle \tag{28}$$

are uniquely determined by the single-spin expectation value $\mathbf{S}_n = \langle \Psi | \hat{\mathbf{S}}_n | \Psi \rangle$. This is trivially true for the nearest-neighbor correlation function $C_{n,n+1}^{\text{hom}}$ of a homogeneous one-dimensional system, which as a consequence of the definition of the homogeneous Hamiltonian (1) is simply given by

$$C_{n,n+1}^{\text{hom}} = \frac{1}{NJ} E_0(S), \tag{29}$$

but the above proof guarantees that the more complicated correlation functions involving more than two spins and/or spatially inhomogeneous spin distributions are, in principle, also functions of $\mathbf{S}_i$ only.

B. Local-density approximation

Another consequence of this Heisenberg-model formulation of the Hohenberg-Kohn theorem is that the model's ground-state energy and spin distribution can be obtained by application of the variational principle to spin distributions, instead of wave functions. In complex situations this can be a major simplification, but to extract this information is, of course, still highly nontrivial. The most straightforward thing to do would be to set up an approximation for the total energy of the system under study as a functional of the spin distribution, and to minimize with respect to $\mathbf{S}_i$. In *ab initio* DFT this is not the preferred way to proceed because it turns out to be hard to conceive good density functionals for the kinetic energy. In practical applications of *ab initio DFT* one therefore commonly employs an indirect minimization scheme, leading to the widely used Kohn-Sham equations. Although this could also be done in the present case, there is no need for introducing a Kohn-Sham system for the Heisenberg model, since there is no kinetic-energy term in the first place. Direct minimization of total-energy expressions seems much more convenient (and more analogous to the way the model is usually treated in statistical physics) than indirect minimizations. In order to explore how such a direct minimization can proceed we first construct, in this subsection, a local-density approximation for the simpler Heisenberg models discussed in the preceding section.

Let us write the two contributions to the ground-state energy of the inhomogeneous Heisenberg model as E_J and E_B, which are the ground-state expectation values of the first and second terms on the right-hand side of Eq. (2), respectively. The mean-field approximation for E_J yields

$$E_0^{\mathrm{MF}}[\mathbf{S}_i] = J \sum_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_i \mathbf{S}_i \cdot \mathbf{B}_i \equiv E_J^{\mathrm{MF}}[\mathbf{S}_i] + E_B[\mathbf{S}_i],$$
(30)

where, as above, $\mathbf{S}_i = \langle \Psi | \hat{\mathbf{S}}_i | \Psi \rangle$. In Sec. IV we quantify the error made by the neglect of correlation effects arising from use of $E_J^{\mathrm{MF}}[\mathbf{S}_i]$ in place of $E_J[\mathbf{S}_i]$.

A guideline for the construction of better functionals than $E_J^{\mathrm{MF}}[\mathbf{S}_i]$ is provided by *ab initio* DFT (Ref. 16) or recent work on the Hubbard model.[20–22] In the former, the total energy of an arbitrarily inhomogeneous system is written as

$$E_0[n(\mathbf{r})] = T_s[n(\mathbf{r})] + E_H[n(\mathbf{r})] + E_v[n(\mathbf{r})] + E_{\mathrm{xc}}[n(\mathbf{r})],$$
(31)

where T_s is the noninteracting kinetic energy,

$$E_H[n(\mathbf{r})] = \frac{1}{2} \int d^3 r \int d^3 r' \frac{n(\mathbf{r}) n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}$$
(32)

the Hartree energy, and E_v the potential energy arising from the external field $v(\mathbf{r})$. The local-density approximation (LDA) for the exchange-correlation (xc) energy is

$$E_{\mathrm{xc}}[n(\mathbf{r})] \approx E_{\mathrm{xc}}^{\mathrm{LDA}}[n(\mathbf{r})] = \int d^3 r\, e_{\mathrm{xc}}(n)|_{n \to n(\mathbf{r})}.$$
(33)

This expression locally substitutes the *xc* energy of the inhomogeneous system by the one of a homogeneous system of same density. The necessary input expression for $e_{\mathrm{xc}}(n)$ is obtained by subtracting the Hartree and noninteracting kinetic energy from the ground-state energy of the homogeneous system $e_0(n)$.

In the present context we write, in analogy to Eq. (31),

$$E_0[\mathbf{S}_i] = E_J^{\mathrm{MF}}[\mathbf{S}_i] + E_B[\mathbf{S}_i] + E_c[\mathbf{S}_i],$$
(34)

where E_J^{MF} is defined in Eq. (30), E_B is the potential energy arising from the external field $\mathbf{B}_i$, and E_c is by definition the difference between the mean-field result and the correct one, i.e., the correlation energy. (There is no Heisenberg-model counterpart to the kinetic energy term, and we avoid the expression "exchange-correlation energy" because in common terminology the entire Heisenberg Hamiltonian is due to "exchange.")

To obtain an explicit scheme we now propose to approximate, in analogy to Eq. (33),

$$E_c[\mathbf{S}_i] \approx E_c^{\mathrm{LDA}}[\mathbf{S}_i] = \sum_i e_c(S)|_{S \to |\mathbf{S}_i|},$$
(35)

where $e_c(S)$ is obtained by subtracting the mean-field energy $-dS^2$ from the homogeneous expressions for $e_0(S)$ discussed in Sec. II. As an explicit example, the LDA approxi-

mation for the correlation energy of an *inhomogeneous* antiferromagnetic Heisenberg model in one dimension becomes

$$E_{c,\mathrm{AFM},d=1}^{\mathrm{LDA}}[\mathbf{S}_i] = J\left(\frac{2}{\pi} - 1\right) \sum_i |\mathbf{S}_i|,$$
(36)

where we used Eq. (10) for $e_0(S)$. Of course Eqs. (14) or (15) can be used in the same way in $d=1$, and Eqs. (12) and (13) in $d=2$ and $d=3$, respectively. The full ground-state energy is then for any d approximated as

$$E_0[\mathbf{S}_i] \approx E_0^{\mathrm{LDA}}[\mathbf{S}_i] = E_J^{\mathrm{MF}}[\mathbf{S}_i] + E_B[\mathbf{S}_i] + E_c^{\mathrm{LDA}}[\mathbf{S}_i].$$
(37)

Clearly Eq. (35) is a rather simple approximation, whose quality may vary widely depending on the circumstances (e.g., values of J and d, or spatial dependence of $\mathbf{B}_i$). At present it is motivated mainly by the considerable practical success of its counterpart in *ab inito* DFT,[16–18] and by the encouraging results obtained recently with a Bethe-ansatz based LDA for inhomogeneous Hubbard models.[20–22] It is clear, however, that the LDA contains essential correlation effects not accounted for by the mean-field expression (30). In spite of the extra term, minimization of Eq. (34) with (35) is no more complicated than that of Eq. (30). Equation (35) thus shows one way in which the expressions listed in Sec. II for homogeneous systems can be applied to inhomogeneous situations. A simple application of these ideas is worked out in Sec. IV B.

C. A scaling hypothesis

An interesting feature of the functionals obtained by combining Eq. (35) with the explicit formulas of Sec. II is that the resulting expressions depend explicitly on the interaction J and the dimensionality d. The dependence of the *ab initio* functionals on the interaction and the dimensionality is not well known, and in particular the d dependence of the functional is still subject of many ongoing investigations.[30] Even in the much simpler case of the Hubbard model it is only the interaction dependence which is featured explicitly in the available functionals,[19–22] whereas the dependence on dimensionality is essentially unknown. In this context it may be useful to have, for the Heisenberg model, a number of simple expressions that depend explicitly on dimensionality and interaction, so that the role of these parameters in the functional can be explored in a simplified environment.

As an explicit example, we consider scaling properties of the functional as a function of dimensionality d. The Hartree-like term $\propto S^2$ in Eqs. (11) to (13) clearly scales linearly with d. Hence

$$e_J^{\mathrm{MF}}(d) = d e_J^{\mathrm{MF}}(d=1).$$
(38)

Interestingly, the correlation energy contribution $\propto S$ also obeys a similar, albeit less obvious, scaling law. From the explicit expressions (11)–(13) for $e_0^{\mathrm{AFM}}(d)$ one obtains for the relation between the ratio of the correlation energies and the ratio of the dimensionalities

$$\frac{e_c(d=2)}{e_c(d=1)} = 0.870 = \left(\frac{2}{1}\right)^x \qquad (39)$$

and

$$\frac{e_c(d=3)}{e_c(d=2)} = 0.921 = \left(\frac{3}{2}\right)^y, \qquad (40)$$

where x and y are exponents to be determined. Numerically one finds $x = -0.201$ and $y = -0.203$. The near equality of these two exponents among each other and to the integer fraction $-1/5$ leads us to conjecture the following dimensional scaling law:

$$e_c(d) = d^{-\eta} e_c(d=1), \qquad (41)$$

where the scaling exponent $\eta = 1/5$. This scaling law accounts for the numbers in Eqs. (11)–(13) to within $\approx 10^{-3}$.

Of course, at present the scaling law (41) is only a conjecture, but one that is consistent with the numbers of spin-wave theory. It also correctly predicts that as $d \to \infty$ the correlation energy vanishes, leaving behind only the mean-field contribution to the total energy. Since very little is known about the dimension dependence of density functionals we cannot say at present whether the existence of such a law is a mere coincidence, a particular property of the Heisenberg model, or a general phenomenon, but we hope that our observation of dimensional scaling stimulates further research along these lines.

One practical use that can be made of Eq. (41) is to convert an approximate functional obtained for some value of d into one for another dimensionality. Counterparts to this property for other Hamiltonians are interesting not only for *ab initio* calculations (in which many results are known for $d=3$, but much less in $d=2$ or $d=1$),[30] but also in the case of the Hubbard model, in which the LDA functional is known only for $d=1$.[20–22]

IV. CORRELATION ENERGY OF THE ANTIFERROMAGNETIC HEISENBERG MODEL

In this section we apply the results obtained above to a study of the correlation energy of the antiferromagnetic Heisenberg model. In Sec. IV A we compare the homogeneous expressions of Sec. II with their mean-field approximation, to assess the importance and behavior of the correlation energy. In Sec. IV B we study a physically interesting inhomogeneity, a spin-density wave, with the LDA functional (36).

A. Homogeneous system

As in the previous section we define the correlation energy as the difference between the total ground-state energy and its mean-field approximation. For a homogeneous system on a linear, square or cubic lattice the latter yields

$$E_0^{\text{MF}}(S, J, d) = -JdNS^2 \qquad (42)$$

and thus $e_0(S) = -dS^2$. In the inset of Fig. 4 we plot the difference between this value and the total energy expres-
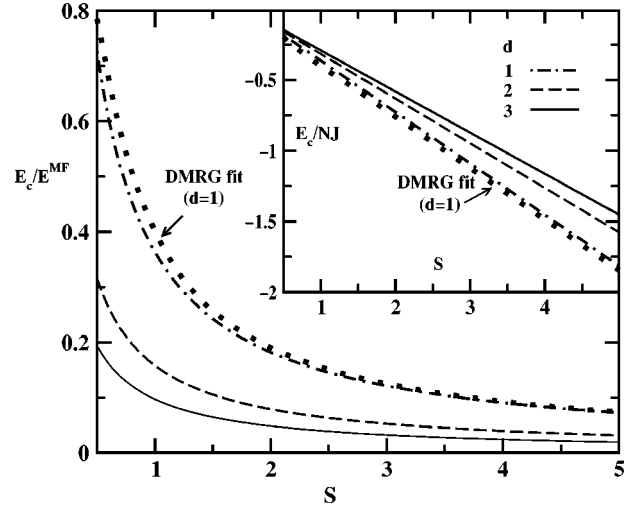

FIG. 4. Correlation energy of the antiferromagnetic homogeneous Heisenberg model in one, two, and three dimensions, as a function of spin. The curves labeled $d = 1,2,3$ are obtained by subtracting the mean-field energy from the expressions (11)–(13), and the one labeled "$d = 1$ DMRG fit" is obtained by using our fit Eq. (15) instead of Eq. (11). Main figure: correlation energy relative to mean-field energy. Inset: absolute size of correlation energy.

sions Eqs. (11), (12), and (13). For one dimension we also plot the difference between the mean-field result and the more precise expression (15). These differences represent the *absolute* size of the correlation energy. In the main part of Fig. 4 we display the *relative* size of the correlation energy as compared to the mean-field energy. Several conclusions can be drawn from inspection of these curves.

(i) The inset of Fig. 4 shows that the absolute size of the correlation energy increases towards larger spins. This seems counterintuitive, because larger spins should more closely mimic the classical limit, in which there are no quantum fluctuations and the mean-field approximation becomes exact in the ground state. However, as shown in the main figure, the *relative* weight of the correlation energy as compared to the mean-field energy decreases towards larger spins. Interestingly, the naive expectation that correlations should become less important near the classical limit is thus only true in relative terms, but not in absolute ones.

(ii) On similar grounds one would expect that correlations become less important for larger dimensionality. This is confirmed both by the main figure and the inset, showing that correlations decrease in absolute size and relative to the mean-field energy as d increases. The way the d dependence approaches the classical limit is thus qualitatively different from the way the spin dependence does.

(iii) The mean-field energy is not reliable for any dimensionality $d \leq 3$ and $S < 5$, leading to errors that can be larger than J in absolute size and larger than 50% in relative terms. This observation puts tight limits on the reliability of this rather widely used approximation.

(iv) The improved treatment of correlations in $d = 1$, represented by the curves labeled "DMRG fit," does not invalidate conclusions (i), (ii), and (iii), obtained on the basis of the spin-wave expressions (11)–(13). However, both the

main figure and the inset of Fig. 4 show that it enhances the importance of correlations with respect to the mean-field values, as compared to the simpler expressions.

B. Example of an inhomogeneous system: a linear spin-density wave

As an example of a truly inhomogeneous situation, to which our LDA functional (35) can be applied, we now consider a simple but physically interesting inhomogeneity, namely, a spin-density wave (SDW) imposed by an external field on a chain with antiferromagnetic coupling.

We model the SDW state by taking

$$\mathbf{S}_n = S\mathbf{u}_x \cos\phi_n, \tag{43}$$

where $\phi_n = 2\pi(n-1)/\lambda$ and $\mathbf{u}_x$ denotes the unit vector in the x direction. This choice describes a linear SDW of amplitude S and wave length λ, polarized along the x direction. (The lattice is taken to be a chain along the z direction.) The corresponding mean-field energy is

$$E_0^{\mathrm{MF}}[\mathbf{S}_i] = J\sum_{n=1}^{N-1} \mathbf{S}_n \cdot \mathbf{S}_{n+1} + \sum_{n=1}^{N} \mathbf{S}_n \cdot \mathbf{B}_n \equiv E_J^{\mathrm{MF}}[\mathbf{S}_i] + E_B[\mathbf{S}_i], \tag{44}$$

where N is the number of lattice sites and $\mathbf{B}_n$ is a magnetic field that can be thought of as either externally applied [thus forcing the system into a state with spin distribution (43)] or generated self-consistently, or a combination of both. The LDA approximation for the ground-state energy is, on the other hand,

$$E_0^{\mathrm{LDA}}[\mathbf{S}_i] = E_0^{\mathrm{MF}}[\mathbf{S}_i] + E_c^{\mathrm{LDA}}[\mathbf{S}_i] = E_J^{\mathrm{MF}}[\mathbf{S}_i] + E_B[\mathbf{S}_i]$$
$$+ \sum_{n=1}^{N} e_c^{\mathrm{AFM}}(S)\big|_{S\to|\mathbf{s}_i|} \equiv E_J^{\mathrm{LDA}}[\mathbf{S}_i] + E_B[\mathbf{S}_i], \tag{45}$$

where we use, for simplicity, Eq. (36) for $E_c^{\mathrm{LDA}}[\mathbf{S}_i]$.

For the given spin distribution (43) we now compare the predictions of the mean-field and LDA expressions for the interaction energy E_J. Note that this is not a self-consistent calculation, but a comparison of the two expressions (44) and (45) for a fixed distribution specified by Eq. (43). In Fig. 5 we plot E_J^{MF}, E_J^{LDA}, and E_c^{LDA} as functions of λ, the wave length of the SDW. Since the term E_B, arising from the magnetic field, is the same in both approximations we only display the interaction energy E_J. The presence of an external field with symmetry different from the one of the ground-state of the unperturbed antiferromagnetic system gives rise to a rich physics. Particularly, we note the following points.

(i) Addition of the correlation energy to the mean-field result lowers the total energy considerably. This is, qualitatively, the same behavior we found previously in homogeneous systems and illustrates again the importance of going beyond the mean-field expression for the energy.

(ii) As $\lambda \to \infty$ the SDW approaches a ferromagnetic spin configuration. Since the unperturbed homogeneous model is antiferromagnetic this state is energetically unfavored, and
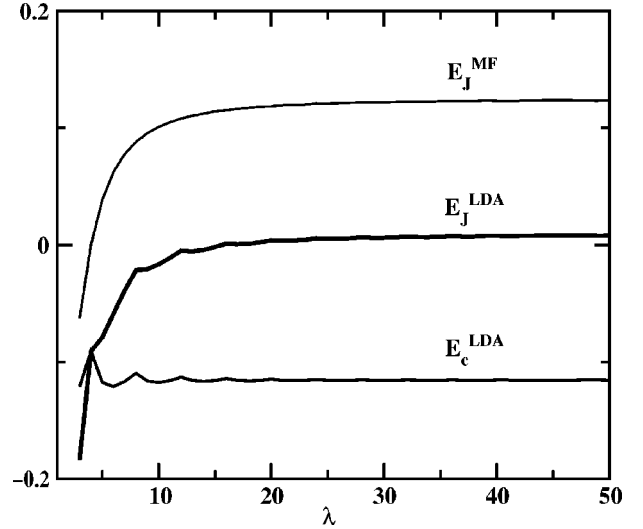

FIG. 5. Interaction energy E_J of a spin-density wave imposed on an antiferromagnetic chain, as a function of wave length of the spin modulation. Upper curve: mean-field approximation, defined by Eq. (44). Lower curve: local-density approximation for the correlation energy, as given by Eq. (36). Middle curve: LDA result for interaction energy, obtained as sum of the two other curves.

corresponds to a maximum of the E versus λ curves. In the opposite limit the spatial modulation of the SDW can take local advantage of the AFM tendency of the underlying homogeneous system. This leads to a lowering of the energy as $\lambda \to 0$.

(iii) Once λ is larger than approximately 10 lattice constants the E versus λ curves saturate. We interpret this in terms of the correlation length of the antiferromagnetic model by noting that once the SDW modulation takes place on a scale larger than a few correlation lengths, the system will be relatively insensitive to further approximation to the ferromagnetic state.

(iv) The overall downshift of the LDA curve compared to the mean-field one implies that the ferromagnetic configuration is energetically less unfavorable in the former approximation than in the latter. Physically this is reasonable, because the correlations accounted for by E_c break up the rigid AFM pattern of the Néel state found in the mean-field approximation, and replace it by a complex ground state involving spins along all three directions in space.

V. SUMMARY AND OUTLOOK

Density-functional theory is commonly applied to the *ab initio* Hamiltonian, in the context of electronic structure calculations of molecules or solids.[16-18] Applications to model Hamiltonians are rare, although they can be useful both for the analysis of these models in the presence of inhomogeneity (broken translational invariance), and for further development of DFT. The present work on the Heisenberg model serves to exemplify these two complementary aspects.

The Heisenberg Hamiltonian is much simpler than the Hubbard Hamiltonian, which describes the charge degrees of freedom in addition to the spin ones, or than the *ab initio*

Hamiltonian involving the long-range Coulomb interaction between charges in a real crystal. The Heisenberg model may therefore be considered a simplified environment in which concepts and methods of DFT can be tested and analyzed. An interesting example is the study of the interaction and dimension dependence of *xc* functionals, about which little is known in the *ab initio* case.

On the other hand, the LDA for inhomogeneous Heisenberg models is no more complicated, formally, than the mean-field approximation, but it accounts by construction for essential correlation effects missed by the latter. Analysis of the correlation energy of homogeneous and inhomogeneous Heisenberg models, as function of spin and dimensionality, illustrates that such effects are crucial for a quantitative description of the ground state. The above LDA functional may thus be useful in studies of the behavior of the Heisenberg model in spatially varying external fields or in the presence of internal magnetic order that breaks translational symmetry.

The simple LDA-type correlation functional based on Eqs. (35) and (11) is seen to produce significant quantitative change and qualitative improvement over the mean-field approximation, at very little extra computational cost. This observation encourages us to envisage more complex applications of this functional, such as to impurity states in the Heisenberg model. An extension of the present work to a study of the thermodynamics of inhomogeneous Heisenberg models [employing the $T>0$ formulation of DFT (Ref. 31)] is also planned for the future.

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