Relationship between the wave function of a magnet and its static structure factor

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We state and prove two theorems about the ground state of magnetic systems described by very general Heisenberg-type models and discuss their implications for magnetic neutron scattering. The first theorem states that two models cannot have the same correlator without sharing the corresponding ground states. According to the second theorem, an N-qubit wave function cannot reproduce the correlators of a given system unless it represents a true ground state of that system. We discuss the implications for neutron scattering inverse problems. We argue that the first theorem provides a framework to understand neutron-based Hamiltonian learning. Furthermore, we propose a variational approach to quantum magnets based on the second theorem in which a representation of the wave function (held, for instance, in a neural network or in the qubit register of a quantum processor) is optimized to fit experimental neutron scattering data directly, without the involvement of a model Hamiltonian.

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I. INTRODUCTION

The Rayleigh-Ritz variational principle states that the ground-state wave function of a quantum system is an absolute minimum of the energy. It provides the theoretical underpinning of many successful approaches to the quantum many-body problem, including density functional theory (DFT) [1], variational Monte Carlo methods [2], the BCS theory of superconductors [3], and the Laughlin theory of the fractional quantum Hall effect [4], to name a few cases. More recently, it has been used to find optimal representations of wave functions using quantum computers [5,6] and neural networks [7]. Such theories start with a model Hamiltonian $\hat{H}$ and proceed by minimizing the energy $\langle\Psi|\hat{H}|\Psi\rangle$ to obtain the wave function Ψ. The Rayleigh-Ritz variational principle ensures that no wave function can yield a lower value of the energy than the system's true ground state. Once the wave function is known, it is straightforward to predict expectation values of observables. Very often, however, $\hat{H}$ is not known *a priori*. In such instances $\hat{H}$ has to be found from experimental data. That involves a laborious and ill-posed inverse problem: multiple candidate Hamiltonians must be studied until one is found that predicts the experimentally determined value of a set of observables. In general there is no guarantee of uniqueness of $\hat{H}$ or Ψ for a given data set. Here we consider the inverse problem for the magnetic structure factor of a magnetic insulator (in particular, one described by an anisotropic Heisenberg model, which covers a vast range of real materials). We show that, for systems that have nondegenerate, distinct ground states, there is a one-to-one correspondence between the structure factors, the model Hamiltonian, and the ground-state wave function. We then address the implications of degeneracy, Hamiltonians with the same ground state, and excitations and discuss the implications for neutron scattering.

Our results have several direct implications for the study of magnetic insulators using neutron scattering, specifically for the neutron scattering inverse problems described schematically in Fig. 1. First, as we argue below, Theorem 1 puts the Hamiltonian-learning problem [Fig. 1(a)] on firmer footing and will help the design of efficient solutions, for instance, ones exploiting machine learning [8]. Second, Theorem 2 suggests, and supports, variational methods in which the wave function is optimized to describe the experimental data, obviating the need to minimize the energy of a model Hamiltonian [Fig. 1(b)]. This provides an alternative to existing methods used to obtain the ground state of a Heisenberg-type magnet, for instance, those based on neural-network [7] or quantum-processor [5] representations. Many such methods are based on minimizing the energy for a given model. The methods discussed in this work will be appropriate when the model is not yet known but experimental structure factor information is available. In those circumstances, working with the wave function directly has the advantage of involving a single optimization loop rather than two nested ones [compare Figs. 1(a) and 1(b)]. In analogy with the Rayleigh-Ritz variational principle, our second theorem guarantees that no wave function other than a true ground-state wave function of the system under investigation can yield a better fit to the data. Finally, our results suggest that every ground-state property of the system is contained in the structure factors. This has important implications for efforts to quantify quantum entanglement from experimental neutron scattering data [9–11] and justifies the reduction of measures of entanglement to functions of correlators [12,13].

The work presented here has to be seen in the context of recently developed methods for the determination of model Hamiltonians from local measurements [14–18]. Interestingly, a main thrust of such works, which are usually concerned with systems in which qubits need to be addressed

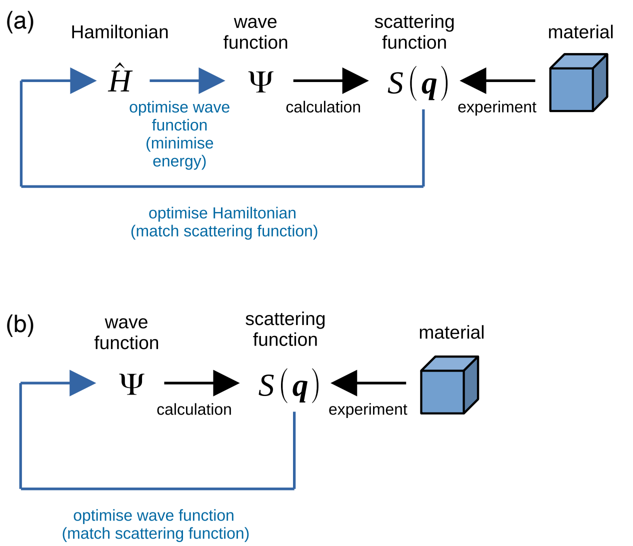

FIG. 1. Two versions of the diffuse magnetic neutron scattering inverse problem: (a) given the scattering function $S(\mathbf{q})$ of a real material, in Hamiltonian learning the aim is to determine a model Hamiltonian $\hat{H}$ whose wave function Ψ will describe $S(\mathbf{q})$ satisfactorily. (b) In quantum tomography one tries to determine the wave function Ψ directly.

individually, is the optimization of the scaling of the number and type of measurements required with the size of the system and the range of interactions. In contrast, our approach relies on the static magnetic structure factor $S_{\alpha,\beta}(\mathbf{q})$, which contains information about all two-point correlators and can be determined experimentally with the same effort irrespective of system size or range of interactions.

II. THEOREMS

Our starting assumption is that the physical system under experimental investigation can be described by an anisotropic Heisenberg model:

$$\hat{H} = \sum_{i,j} \sum_{\alpha,\beta} J_{i,j}^{\alpha,\beta} \hat{S}_i^\alpha \hat{S}_j^\beta. \tag{1}$$

Here $i, j = 1, 2, \ldots, N$ represent atomic sites whose positions $\mathbf{R}_i$ and $\mathbf{R}_j$ we assume are known. $\hat{S}_i^\alpha$ represents the αth component of the spin operator for the magnetic moment at the ith atomic site ($\alpha = x, y, z$; we assume each spin component is defined with reference to some local axes defined on each site). We assume the spin quantum number at each site is $S = 1/2$ in what follows, but the results can be generalized to arbitrary S straightforwardly. $J_{i,j}^{\alpha,\beta}$ is an exchange constant describing the interaction between the αth component of the spin at the ith site of a given lattice and the β component of the spin in the jth site. The terms with $i = j$ describe site anisotropy (e.g., easy planes or easy axes). The dependence of $J_{i,j}^{\alpha,\beta}$ on i, j, α, and β is entirely arbitrary. The model in Eq. (1) can thus describe a very broad range of magnetic models in arbitrary dimensions with and without translational invariance, including the Ising model [19], XY model [20], and Kitaev model, to name but a few [21]. Models of this type are believed to describe well the physics of many materials from single-molecule magnets [22] through infinite-chain compounds [11] to three-dimensional quantum spin ices [23] and other spin liquids [24]. The observable quantity of interest is the two-point magnetic correlator

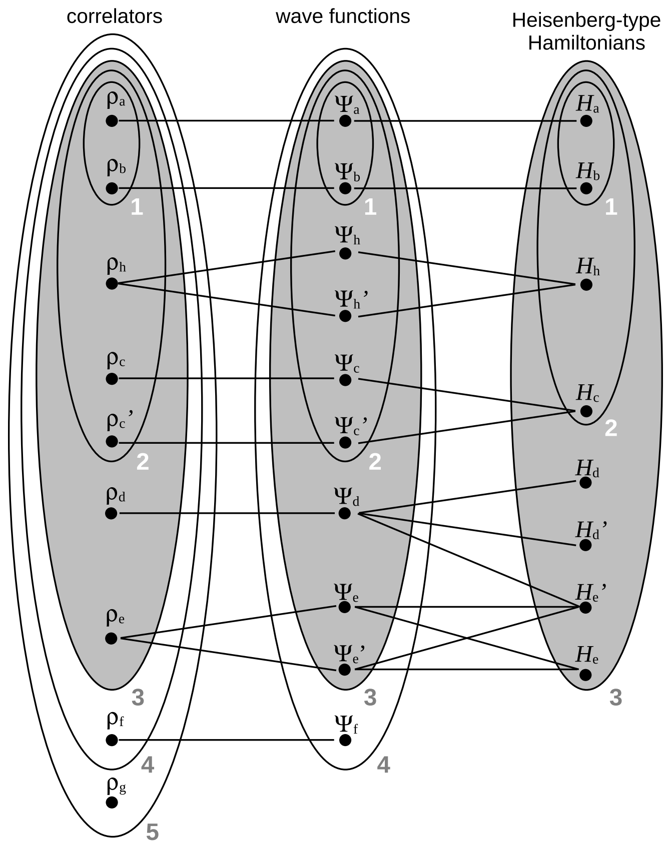

FIG. 2. Schematic illustration of possible relationships between two-point correlation functions ρ, N-qubit wave functions Ψ, and general spin-1/2 anisotropic Heisenberg Hamiltonians H (see the text).

$$\rho_{i,j}^{\alpha,\beta}[\Psi] \equiv \langle \Psi | \hat{S}_i^\alpha \hat{S}_j^\beta | \Psi \rangle. \tag{2}$$

The correlator is obviously a single-valued functional of the wave function Ψ. As shown in Appendix A, this quantity is readily obtainable in condensed-matter systems through neutron scattering measurements of the static structure factor $S_{\alpha,\beta}(\mathbf{q})$. The situation we have in mind is one in which the ground-state correlator $\rho_{i,j}^{\alpha,\beta}[\Psi_0]$ has been obtained experimentally but neither the Hamiltonian $\hat{H}$ nor the wave function Ψ_0 is known. We wish to prove two closely related theorems that impose constraints on $\hat{H}$ and Ψ_0:

Theorem 1. Two Hamiltonians $\hat{H}$ and $\hat{H}'$ cannot have the same ground-state correlator without sharing the corresponding ground states.

Theorem 2. Any N-qubit wave function Ψ that can reproduce the ground-state correlator of $\hat{H}$ represents a ground state of $\hat{H}$.

The implications of these two theorems for the relationship between correlators, wave functions, and Hamiltonians are illustrated in Fig. 2. For nondegenerate Hamiltonians, Theorem 1 is a particular case of a more general theorem proven by Ng [25]. Here we extend it to the important case of Hamiltonians with degenerate ground states and discuss the implications of Hamiltonians sharing a ground state for Hamiltonian learning using neutron scattering data [Fig. 1(a)]. The latter discussion will be supported by a generalization of Theorem 1 to excited states (Appendix B). Theorem 2, on the other hand, is a consequence of the fact that the ground state maximizes the reconstruction entropy given a set of local measurements [26]. The theorems are also closely related to the convexity of the set of all two-point correlators of N-qubit states [27,28]. Note that our second theorem does not require all correlators to be representable by N-qubit wave functions (exemplified by $\rho_{\rm g}$ Fig. 2), nor is it restricted to trial wave functions that are ground states of Heisenberg-type Hamiltonians ($\rho_{\rm f}$ in Fig. 2). We will discuss the implications of this second theorem for neutron-based quantum tomography [Fig. 1(b)]. Finally, we note that Theorem 1 can be deduced from Theorem 2; however, for clarity we will prove both theorems independently.

Our approach to proving Theorems 1 and 2 is inspired by the DFT formalism for Heisenberg models developed by Líbero and Capelle [29]. Our aims, however, are quite different. The latter work (like other DFT formalisms for lattice models [29–31]) is an energy-minimization variational theory closely modeled on the original DFT for electrons in solids [1]. In density functional theories generally, the aim is to show that the energy is a functional of a densitylike quantity (in the case of Ref. [29], the local magnetization). One then splits the energy into two parts, one that is "universal" and another that depends on local fields. In order for this to be useful, it is necessary to have exact results for the universal function and motivated approximations for the field-dependent contribution. Here our primary quantity is not a density but a correlator, and we are not interested in splitting the energy into one contribution that is known and another that is to be approximated. Instead, we treat the energy as a single unit and are interested in proving that only one universal Hamiltonian is compatible with a given set of correlators. In practice, applications of our approach involve the optimization of the match to experimental data, rather than the minimization of the energy. Moreover, we will work in the absence of a known model Hamiltonian, rather than using knowledge of one model (e.g., a translationally invariant Heisenberg model) to approximately solve another (e.g., the same model but with an impurity potential).

III. PROOF OF THEOREM 1

Inspired by the original proof of the Hohenberg-Kohn theorem of DFT, we will proceed by *reductio ad absurdum*. Suppose there are two distinct Hamiltonians $\hat{H}$ and $\hat{H}'$, with different exchange interaction functions $J^{\alpha,\beta}_{i,j}$ and $J'^{\alpha,\beta}_{i,j}$ and different ground states $|\Psi_0\rangle, |\Psi_0'\rangle$, respectively, that give the same correlator $\rho^{\alpha,\beta}_{i,j}$:

$$\rho^{\alpha,\beta}_{i,j}[\Psi_0] = \rho^{\alpha,\beta}_{i,j}[\Psi_0'] \text{ for all } i, j, \alpha, \beta. \tag{3}$$

We will first consider the case when the two ground states are nondegenerate. In this case the ground-state energy obtained from the first Hamiltonian is

$$\begin{aligned} E_0 &= \langle\Psi_0|\hat{H}|\Psi_0\rangle < \langle\Psi_0'|\hat{H}|\Psi_0'\rangle && (4)\\ &= \langle\Psi_0'|\hat{H}-\hat{H}'|\Psi_0'\rangle + \langle\Psi_0'|\hat{H}'|\Psi_0'\rangle && (5)\\ &= \sum_{i,j}\sum_{\alpha,\beta}\big(J^{\alpha,\beta}_{i,j} - J'^{\alpha,\beta}_{i,j}\big)\rho^{\alpha,\beta}_{i,j}[\Psi_0'] + E_0', && (6)\end{aligned}$$

where the inequality is due to the Rayleigh-Ritz variational principle. Similarly, the ground-state energy obtained from the second Hamiltonian is

$$\begin{aligned} E_0' &= \langle\Psi_0'|\hat{H}'|\Psi_0'\rangle < \langle\Psi_0|\hat{H}'|\Psi_0\rangle && (7)\\ &= \langle\Psi_0|\hat{H}'-\hat{H}|\Psi_0\rangle + \langle\Psi_0|\hat{H}|\Psi_0\rangle && (8)\\ &= \sum_{i,j}\sum_{\alpha,\beta}\big(J'^{\alpha,\beta}_{i,j} - J^{\alpha,\beta}_{i,j}\big)\rho^{\alpha,\beta}_{i,j}[\Psi_0] + E_0. && (9)\end{aligned}$$

Adding the two inequalities, we obtain

$$E_0' + E_0 < \sum_{i,j}\sum_{\alpha,\beta}\big(J'^{\alpha,\beta}_{i,j} - J^{\alpha,\beta}_{i,j}\big) \times \big\{\rho^{\alpha,\beta}_{i,j}[\Psi_0] - \rho^{\alpha,\beta}_{i,j}[\Psi_0']\big\} + E_0 + E_0'.$$

Using now our assumption (3), this reduces to

$$E_0' + E_0 < E_0 + E_0', \tag{10}$$

which is absurd. Thus, our initial assumption must be incorrect: two Heisenberg-type Hamiltonians with different exchange interaction constants and distinct, nondegenerate ground states can never give the same correlator. In other words, for nondegenerate Hamiltonians that do not share ground states the exchange interaction function is a single-valued functional $J^{\alpha,\beta}_{i,j}[\rho]$ of the correlator $\rho^{\alpha,\beta}_{i,j}$. This is illustrated by the one-to-one correspondence between $\{H_{\rm a}, H_{\rm b}\}$ and $\{\rho_{\rm a}, \rho_{\rm b}\}$ in Fig. 2.

We note that our proof relies on the assumption that $|\Psi_0\rangle \neq |\Psi'{}_0\rangle$ since otherwise, the strict inequalities (4) and (7) become equalities. In other words, if $\hat{H}$ and $\hat{H}'$ share their unique ground state, the theorem does not apply. This is illustrated by the one-to-many correspondence between $\rho_{\rm d}$ and $\{H_{\rm d}, H_{\rm d}', H_{\rm e}'\}$ in Fig. 2. Although this might appear to be a serious limitation for Hamiltonian learning using neutron scattering, it may not be as important in practice, as we discuss below.

In the above paragraphs we explicitly assumed that the ground states of $\hat{H}$ and $\hat{H}'$ are nondegenerate. In order to prove Theorem 1 we need to relax that assumption. Let us first consider the case when the ground state of one of the Hamiltonians (which we take to be $\hat{H}$ without loss of generality) is degenerate while that of the other Hamiltonian remains nondegenerate. Then the first of the above two inequalities (4) and (7) is not strict, as there is always the possibility that $|\Psi_0'\rangle$ happens to be a ground state of $\hat{H}$ as well as being the unique ground state of $\hat{H}'$. In that case, Theorem 1 would be violated because $|\Psi_0\rangle$ would not be a ground state of $\hat{H}'$ but it would have the same correlators as $|\Psi_0'\rangle$, which is. Barring that possibility, the arguments above hold, so we need to consider only that special case. In the special case we have

$$E_0 = \sum_{i,j}\sum_{\alpha,\beta}\big(J^{\alpha,\beta}_{i,j} - J'^{\alpha,\beta}_{i,j}\big)\rho^{\alpha,\beta}_{i,j}[\Psi_0'] + E_0' \tag{11}$$

and

$$E_0' < \sum_{i,j}\sum_{\alpha,\beta}\left(J'^{\alpha,\beta}_{i,j} - J^{\alpha,\beta}_{i,j}\right)\rho^{\alpha,\beta}_{i,j}[\Psi_0] + E_0. \tag{12}$$

When we add the equality (11) to this inequality (12), we still arrive at the same contradiction as before, (10). This proves that Theorem 1 also applies when one of the two Hamiltonians is degenerate.

Let us now consider the case when *both* Hamiltonians have ground-state degeneracy. Then there is a new possibility, namely, that $|\Psi_0\rangle$ is a ground state of $\hat{H}'$ *and* $|\Psi_0'\rangle$ is a ground state of $\hat{H}$ (all other possibilities have already been covered above). In that case all the above inequalities become equalities, and we do not arrive at a contradiction. However, in this case all the ground states of $\hat{H}$ and $\hat{H}'$ leading to the same correlators are shared, so the premise of the theorem is not satisfied. In summary, degenerate Hamiltonians with the same correlators must share *all* the corresponding ground states, as a single ground state of one which is not also a ground state of the other suffices to generate a contradiction. This is illustrated by ρ_e in Fig. 2: the same correlator can be generated by the ground state of H_e and by that of H_e', but then the ground state of H_e' must also be a ground state of H_e and *vice versa* (it is possible, on the other hand, for ground states leading to *different* correlators not to be shared, as illustrated by the correlator ρ_d of H_e'). This concludes our proof of Theorem 1. ■

IV. HAMILTONIAN LEARNING

Before proving Theorem 2 we shall discuss the implications of Theorem 1 for Hamiltonian learning using neutron scattering. First of all, we address the question of shared ground states. An example of this would be two spin-1/2 ferromagnetic Ising models differing only by an overall multiplicative factor. In both cases, the ground state is a classical state where all the spins point along the positive or negative direction of the quantization axis. The correlators are therefore identical in the ground state. It is therefore impossible to discriminate between these two models from a measurement of the ground-state correlators. It would be tempting to venture that this limitation of Theorem 1 can be trivially circumvented by normalizing the parameters of trial Hamiltonians. However, this would take care of only certain instances of shared ground states, known *a priori*. We cannot discard nontrivial cases. On the other hand, it is straightforward to generalize our proof of Theorem 1 to show that it holds for any excited state $|\Psi_n\rangle$ as well as for the ground state (see Appendix B). Therefore, two Hamiltonians that are physically distinct but share a ground state could be told apart by probing their low-energy excitations. Indeed, any real condensed-matter experiment will take place at finite temperature with the measured correlator corresponding to a thermal superposition $\sum_n Z^{-1}\exp(-E_n/k_BT)\rho^{\alpha,\beta}_{i,j}[\Psi_n]$. For two Hamiltonians $\hat{H}$ and $\hat{H}'$ to give the same result at any arbitrary temperature one would therefore require all eigenstates $|\Psi_n\rangle$ and all the eigenvalues E_n to coincide. In that case, the two Hamiltonians are identical: $\hat{H} = \hat{H}' = \sum_n E_n|\Psi_n\rangle\langle\Psi_n|$. Thus, this limitation of Theorem 1 may not, in practice, limit its applicability to Hamiltonian learning in neutron scattering experiments [32,33].

The above discussion suggests that the inverse problem of deducing the Hamiltonian from the correlators may, indeed, be well defined, as long as we know that the material under investigation is described by a model of the form in Eq. (1). That could provide a natural explanation for the success of a recent machine learning based approach to this problem [8]. In that reference an autoencoder was trained using simulations of the neutron scattering function $S_{\alpha,\beta}(\mathbf{q})$ obtained for a family of candidate Hamiltonians [for completeness, we offer a proof of the equivalence between knowledge of $S_{\alpha,\beta}(\mathbf{q})$ and of $\rho^{\alpha,\beta}_{i,j}$ in Appendix A]. The autoencoder thus trained can be used to generate a low-dimensional latent space on which experimental data can be projected, effectively finding an optimal model Hamiltonian. Although, in principle, that inverse problem is "ill defined" [8], our formal results for the ground and excited states of Heisenberg-type Hamiltonians strongly suggest that there may be only one solution. We note that the work in Ref. [8] dealt with classical models; however, similar dimensionality reduction has been shown for quantum models using closely related principal component analysis [34].

Several recent works discussed the determination of model Hamiltonians using local measurements [14–18]. While such methods are well suited to artificial systems such as quantum simulators, they are not readily applicable to experimental data on condensed-matter systems. Specifically, in most cases [14–17] they require the covariance matrix, which in turn relies on four-point correlators of the form $\rho^{\alpha,\beta,\alpha',\beta'}_{i,j,i',j'}[\Psi_0] \equiv \langle\Psi_0|\hat{S}^\alpha_i\hat{S}^\beta_j\hat{S}^{\alpha'}_{i'}\hat{S}^{\beta'}_{j'}|\Psi_0\rangle$, where i and i' are sites that are not linked by a direct interaction and j and j' are sites that interact with i and i', respectively. Such higher-order correlators are not readily accessible through neutron scattering. In contrast, for periodic systems all the two-point correlators $\rho^{\alpha,\beta}_{i,j}[\Psi_0]$ can be determined in a neutron scattering experiment (see Appendix A). As an added benefit, such an approach yields all the required information using a "single-shot" global measurement irrespective of the size of the system or the range of spin-spin interactions. Thus, in the case of condensed-matter systems it is not necessary to devise more sophisticated observables in order to improve sampling efficiency, as was proposed recently [18]. Effectively, neutron scattering integrates a large number of local measurements into a single function $S(\mathbf{q})$ that needs to be fitted (see Appendix A)—a sort of analog parallel computation.

V. PROOF OF THEOREM 2

In the preceding paragraphs we proved that, for any set of Heisenberg-type Hamiltonians that do not share ground states, the exchange constants $J^{\alpha,\beta}_{i,j}$ are single-valued functionals of the correlators $\rho^{\alpha,\beta}_{i,j}$ (sets labeled "2" in Fig. 2). With the additional constraint that the Hamiltonians have nondegenerate ground states (sets labeled "1") the ground state $|\Psi_0\rangle$ is in turn fixed by the choice of $J^{\alpha,\beta}_{i,j}$. Thus, in this case $|\Psi_0\rangle$ is also uniquely determined by $\rho^{\alpha,\beta}_{i,j}$. More generally, Theorem 1 implies that the only ground states of

Heisenberg-type Hamiltonians (including Hamiltonians with degenerate and/or shared ground states) that are compatible with the ground-state correlator of a given model are also ground states of that same model. The various possibilities are shown in Fig. 2: each correlator in the shaded area (sets labeled "3") uniquely identifies a nondegenerate ground state ($\rho_{\rm a} \to \Psi_{\rm a}$, $\rho_{\rm b} \to \Psi_{\rm b}$, $\rho_{\rm c} \to \Psi_{\rm c}$, $\rho_{\rm c}' \to \Psi_{\rm c}'$, $\rho_{\rm d} \to \Psi_{\rm d}$) or a set of degenerate ground states of either one Hamiltonian ($\rho_{\rm h} \to \{\Psi_{\rm h}, \Psi_{\rm h}'\}$) or a set of Hamiltonians that share all those states ($\rho_{\rm e} \to \{\Psi_{\rm e}, \Psi_{\rm e}'\}$) [35]. This is essentially the same as Theorem 2 except for the constraint that the trial wave functions must be ground states of Heisenberg-type Hamiltonians. To prove Theorem 2 we need to show that the result holds even without that constraint. In other words, we need to prove that in Fig. 2 there can be no lines linking correlators in the shaded area (set labeled "3" on the left side) to wave functions in the unshaded area (set labeled "4" in the middle). Again, we proceed by *reductio ad absurdum.* Let us assume that there is a state $|\tilde{\Psi}\rangle$ that gives the same correlator as $|\Psi_0\rangle$:

$$\rho_{i,j}^{\alpha,\beta}[\tilde{\Psi}] = \rho_{i,j}^{\alpha,\beta}[\Psi_0] \text{ for all } i, j, \alpha, \beta. \tag{13}$$

Let us further assume that $|\tilde{\Psi}\rangle$ is *not* the ground state of $\hat{H}$. There are two possibilities: either it is the ground state of some other Heisenberg-type Hamiltonian, or it is not the ground state of a Heisenberg Hamiltonian at all. Below we will not assume either case, so our proof will cover both instances. By the Rayleigh-Ritz variational principle, we know that $|\Psi_0\rangle$ gives the absolute minimum of the energy, which implies

$$E_0 \equiv \langle\Psi_0|\hat{H}|\Psi_0\rangle \leqslant \langle\tilde{\Psi}|\hat{H}|\tilde{\Psi}\rangle. \tag{14}$$

Using Eqs. (1) and (2), we can write this as

$$E_0 \equiv \sum_{i,j}\sum_{\alpha,\beta} J_{i,j}^{\alpha,\beta}\rho_{i,j}^{\alpha,\beta}[\Psi_0] \leqslant \sum_{i,j}\sum_{\alpha,\beta} J_{i,j}^{\alpha,\beta}\rho_{i,j}^{\alpha,\beta}[\tilde{\Psi}].$$

Let us now consider separately the two cases when the two expectation values of $\hat{H}$ in Eq. (14) are different and when they are equal. Let us first consider the case when they are different:

$$E_0 \equiv \langle\Psi_0|\hat{H}|\Psi_0\rangle < \langle\tilde{\Psi}|\hat{H}|\tilde{\Psi}\rangle.$$

Then we have

$$\sum_{i,j}\sum_{\alpha,\beta} J_{i,j}^{\alpha,\beta}\rho_{i,j}^{\alpha,\beta}[\Psi_0] < \sum_{i,j}\sum_{\alpha,\beta} J_{i,j}^{\alpha,\beta}\rho_{i,j}^{\alpha,\beta}[\tilde{\Psi}], \tag{15}$$

and from our assumption (13) this reduces to

$$\sum_{i,j}\sum_{\alpha,\beta} J_{i,j}^{\alpha,\beta}\rho_{i,j}^{\alpha,\beta}[\Psi_0] < \sum_{i,j}\sum_{\alpha,\beta} J_{i,j}^{\alpha,\beta}\rho_{i,j}^{\alpha,\beta}[\Psi_0], \tag{16}$$

which is a contradiction. Therefore, the only possibility is that the two expectation values are equal:

$$E_0 \equiv \langle\Psi_0|\hat{H}|\Psi_0\rangle = \langle\tilde{\Psi}|\hat{H}|\tilde{\Psi}\rangle.$$

However, in that case $\langle\tilde{\Psi}|\hat{H}|\tilde{\Psi}\rangle$ is the absolute minimum E_0, and therefore, $|\tilde{\Psi}\rangle$ is a ground state of $\hat{H}$, which contradicts our starting assumption. Thus, we conclude that the only state that reproduces the ground-state correlator of $\hat{H}$ is the actual ground state of $\hat{H}$ (or one of its ground states, if the ground state of $\hat{H}$ happens to be degenerate). ■

We note that our proof of Theorem 2 does not rely on having proved Theorem 1. Theorem 2 is a simple consequence of the fact that the expectation value of any Hamiltonian of the form (1) is a sum of two-point correlators. Thus, if two states $|\Psi_0\rangle$ and $|\tilde{\Psi}\rangle$ give the same correlators, they must give the same expectation value. Therefore, if $|\Psi_0\rangle$ minimizes the energy, $|\tilde{\Psi}\rangle$ does too. We also stress that Theorem 2 is true even when we include candidate wave functions such as $\Psi_{\rm f}$ in Fig. 2 that are not derived from any Hamiltonian of the form (1) (which makes Theorem 2 stronger than Theorem 1). This means that unconstrained searches in wave function space are guaranteed to be able to find the true ground state.

VI. QUANTUM TOMOGRAPHY

Our last result offers the possibility to study systems for which experimental magnetic neutron scattering data are available by working directly with the wave function, without the need for a model Hamiltonian (Fig. 1b). The same efficient encodings of wave functions that have been developed to obtain the ground state of a given model Hamiltonian could be used to find the wave function that matches the experimental data. For instance, one could encode the wave function in a neural network [7], trained once to reproduce the experimental data (instead of minimizing the energy as done in Ref. [7]). Alternatively, a quantum circuit could be optimized to place the qubits in a quantum processor in a state that reproduces the measurements. In this respect, we note that the simulation of inelastic neutron scattering functions of single-molecule magnets using a quantum processor (for known Hamiltonian) has already been successfully demonstrated [36]. The approach we propose would dispense with the model Hamiltonian and instead optimize the scattering function directly. It would be similar to an evolutionary variational eigensolver [6] except that, again, we would not be minimizing the energy of a model Hamiltonian but would be instead optimizing the wave function to describe the experimental data. Both neural networks (or, more generally, tensor networks [37]) and quantum circuits can, in principle, generate any wave function. Our theorem implies that any general-purpose optimization algorithm will converge towards the right ground state (or another ground state of the same model with the same correlators). Specifically, it guarantees that convergence towards an unphysical wave function that reproduces the data is not possible as there are no wave functions that can describe the data and are not valid solutions to the problem at hand. This is akin to the guarantee offered by the Rayleigh-Ritz variational principle that no wave function can give an energy lower than the true ground-state wave function. Once optimized, our neural network or quantum circuit contains all the obtainable information about the system's ground state and can straightforwardly be used to predict any other ground-state property.

VII. FINAL REMARKS

To conclude we note some limitations of Theorem 2. First, it relies on the assumption that the physical system under investigation is described by a Hamiltonian of the form in Eq. (1). Systems with itinerant electrons or with interaction terms involving three or more spins at a time are therefore excluded. The generalization of our results to such systems is left for subsequent work [38]. Second, Theorem 2 establishes the existence of a fitness peak at Ψ_0 but says nothing about its steepness. The peak could be almost a plateau in some cases, which would complicate practical applications. Investigating this for different models provides another focus for future research. Finally, our theorems refer only to the ground state (apart from the generalization of Theorem 1 to excited states in Appendix B). Further generalizations to states of thermodynamic equilibrium and to excited states are left for future work.

Note added. The generalization of Theorem 1 to finite temperatures was discussed recently by Murta and Fernández-Rossier [39].

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APPENDIX A: EQUIVALENCE BETWEEN CORRELATORS AND THE SPIN STRUCTURE FACTOR

Here we show that, for systems with translational symmetry, the correlators $\rho_{ij}^{\alpha,\beta}$ are unique functionals of the diffuse magnetic neutron scattering function or static spin structure factor $S_{\alpha,\beta}(\mathbf{q})$, which can be determined experimentally [40,41] and is given by

$$S_{\alpha,\beta}(\mathbf{q}) \equiv \frac{1}{N\hbar}\sum_{i,j} e^{i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\langle \hat{S}_i^\alpha \hat{S}_j^\beta\rangle. \tag{A1}$$

This is not quite the same as a Fourier transform, in which case we could say straightaway there is a one-to-one correspondence between $S_{\alpha,\beta}(\mathbf{q})$ and $\langle \hat{S}_i^\alpha \hat{S}_j^\beta\rangle$, but almost. Again, let us proceed by *reductio ad absurdum*. First, we assume that there are two different correlation functions that give the same scattering function. Let us designate these two correlation functions as $\rho_{i,j}^{\alpha,\beta}$ and $\tilde{\rho}_{i,j}^{\alpha,\beta}$. Our assumption is that the difference $\Delta_{i,j}^{\alpha,\beta} \equiv \rho_{i,j}^{\alpha,\beta} - \tilde{\rho}_{i,j}^{\alpha,\beta} \neq 0$. Since they give the same scattering function, we have

$$\begin{aligned} S_{\alpha,\beta}(\mathbf{q}) &= \frac{1}{N\hbar}\sum_{i,j} e^{i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\rho_{i,j}^{\alpha,\beta} \\ &= \frac{1}{N\hbar}\sum_{i,j} e^{i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\tilde{\rho}_{i,j}^{\alpha,\beta} \end{aligned}$$

for all $\mathbf{q}, \alpha, \beta$. The last equality implies that

$$\sum_{i,j} e^{i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\Delta_{i,j}^{\alpha,\beta} = 0 \text{ for all } \mathbf{q}, \alpha, \beta. \tag{A2}$$

Suppose that all magnetic sites are equivalent. Then the function $\Delta_{i,j}^{\alpha,\beta} = \Delta^{\alpha,\beta}(\mathbf{R}_i - \mathbf{R}_j)$, and (A2) becomes

$$\sum_{\mathbf{R}} e^{i\mathbf{q}\cdot\mathbf{R}}\Delta^{\alpha,\beta}(\mathbf{R}) = 0 \text{ for all } \mathbf{q}, \alpha, \beta,$$

which evidently implies $\Delta^{\alpha,\beta}(\mathbf{R}) = 0$ for all $\mathbf{R}$ as the Fourier transform of a null function is a null function which contradicts our original assumption, concluding our argument.

Suppose now that the magnetic sites are not equivalent. Nevertheless, as long as we are dealing with a state with translational symmetry, the function $\rho_{i,j}^{\alpha,\beta}$ will have to be periodic. This periodicity can be established experimentally (for instance, by magnetic neutron crystallography), and it is also straightforward to impose it on the wave function; therefore, we can restrict ourselves to the assumption that $\tilde{\rho}_{i,j}^{\alpha,\beta}$ (and therefore also $\Delta_{i,j}^{\alpha,\beta}$) has the same periodicity [42]. In practice this means that we can write the left-hand side of Eq. (A2) in the following form:

$$\begin{aligned} \sum_{i,j} e^{i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\Delta_{i,j}^{\alpha,\beta} &= \mathcal{N}\sum_{i=1}^{M\times\mathcal{N}}\sum_{j=1}^{M} e^{i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)}\Delta_j^{\alpha,\beta}(\mathbf{R}_i) \\ &= \mathcal{N}\sum_{j=1}^{M} e^{-i\mathbf{q}\cdot\mathbf{R}_j} f_j(\mathbf{q}), \end{aligned}$$

with

$$f_j(\mathbf{q}) = \sum_{i=1}^{M\times\mathcal{N}} e^{i\mathbf{q}\cdot\mathbf{R}_i}\Delta_j^{\alpha,\beta}(\mathbf{R}_i).$$

Here $\mathcal{N}$ is the number of magnetic unit cells (repeating units), and M is the number of sites within a unit cell. Thus, the sum over j runs over all the sites in the first unit cell, while the sum over i runs over all the sites in the lattice. For the expression $\sum_{j=1}^{M} e^{-i\mathbf{q}\cdot\mathbf{R}_j} f_j(\mathbf{q})$ to vanish for all $\mathbf{q}$ we must have each of the $f_j(\mathbf{q})$ for $j = 1, 2, \ldots, M$ vanish independently. But $f_j(\mathbf{q})$ is the Fourier transform of $\Delta_j^{\alpha,\beta}(\mathbf{R}_i)$; therefore, $\Delta_j^{\alpha,\beta}(\mathbf{R}_i)$ must vanish too for each $j = 1, 2, \ldots, M$. This means $\Delta_{i,j}^{\alpha,\beta}$ is identically zero, contradicting again our starting assumption.

There is a third possibility; namely, the system may not be periodic. This applies, for example, when there is quenched disorder. In that case the scattering function $S_{\alpha,\beta}(\mathbf{q})$ is averaged over the disorder and is therefore insufficient to determine the real-space correlator. The extent to which $S_{\alpha,\beta}(\mathbf{q})$ constrains the system's ground state in that case should be an interesting subject for future investigations.

APPENDIX B: EXTENSION OF THEOREM 1 TO EXCITED STATES

Here we extend Theorem 1 to excited states. Consider two Hamiltonians $\hat{H}$ and $\hat{H}'$ of the Heisenberg type [Eq. (1)] but with different sets of coupling constants given by $J_{i,j}^{\alpha,\beta}$ and $J'^{\alpha,\beta}_{i,j}$, respectively. Let us assume that the ground-state correlator $\rho_{i,j}^{\alpha,\beta}[\Psi_0]$ is the same. In that case $|\Psi_0\rangle$ is a ground state of both Hamiltonians due to Theorem 1. Let $|\Psi_1\rangle$ and $|\Psi'_1\rangle$ be the first excited states of $\hat{H}$ and $\hat{H}'$, respectively. We wish to prove that if these two states are different, the corresponding correlators are also different, $\rho_{i,j}^{\alpha,\beta}[\Psi_1] \neq \rho_{i,j}^{\alpha,\beta}[\Psi'_1]$. As with all the other proofs in this paper, we proceed by *reductio ad absurdum*. Let us assume that the opposite is true, in other words, $\rho_{i,j}^{\alpha,\beta}[\Psi_1] = \rho_{i,j}^{\alpha,\beta}[\Psi'_1]$. Then

$$E_1 = \langle\Psi_1|\hat{H}|\Psi_1\rangle < \langle\Psi'_1|\hat{H}|\Psi'_1\rangle \tag{B1}$$

$$= \langle\Psi'_1|\hat{H} - \hat{H}'|\Psi'_1\rangle + \langle\Psi'_1|\hat{H}'|\Psi'_1\rangle \tag{B2}$$

$$= \sum_{i,j}\sum_{\alpha,\beta}\left(J_{i,j}^{\alpha,\beta} - J'^{\alpha,\beta}_{i,j}\right)\rho_{i,j}^{\alpha,\beta}[\Psi'_1] + E'_1, \tag{B3}$$

where in writing the inequality we have made use of our assumption that $|\Psi_1\rangle \neq |\Psi'_1\rangle$. We have also used the fact that both $|\Psi_1\rangle$ and $|\Psi'_1\rangle$ are orthogonal to the shared ground state $|\Psi_0\rangle$. Similarly,

$$E'_1 = \langle\Psi'_1|\hat{H}'|\Psi'_1\rangle < \langle\Psi_1|\hat{H}'|\Psi_1\rangle \tag{B4}$$

$$= \langle\Psi_1|\hat{H}' - \hat{H}|\Psi_1\rangle + \langle\Psi_1|\hat{H}|\Psi_1\rangle \tag{B5}$$

$$= \sum_{i,j}\sum_{\alpha,\beta}\left(J'^{\alpha,\beta}_{i,j} - J_{i,j}^{\alpha,\beta}\right)\rho_{i,j}^{\alpha,\beta}[\Psi_1] + E_1, \tag{B6}$$

with the same assumptions made above. Adding the two inequalities, we obtain

$$E_1 + E'_1 < \sum_{i,j}\sum_{\alpha,\beta}\left(J_{i,j}^{\alpha,\beta} - J'^{\alpha,\beta}_{i,j}\right)\left(\rho_{i,j}^{\alpha,\beta}[\Psi'_1] - \rho_{i,j}^{\alpha,\beta}[\Psi_1]\right) + E'_1 + E_1.$$

Our assumption that $\rho_{i,j}^{\alpha,\beta}[\Psi'_1] = \rho_{i,j}^{\alpha,\beta}[\Psi_1]$ then leads to

$$E_1 + E'_1 < E'_1 + E_1,$$

which is absurd. Q.E.D. The argument can be trivially extended to successive excited states. We can also extend it in the same way as Theorem 1 to cover the case where the excited state is degenerate (in other words, to show that if $|\Psi_1\rangle$ and $|\Psi'_1\rangle$ are degenerate excited states of $\hat{H}$, then both of them must also be degenerate states of $\hat{H}'$). ■

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