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Letter

One-to-one correspondence between thermal structure factors and coupling constants of general bilinear Hamiltonians

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A theorem that establishes a one-to-one relation between zero-temperature static spin-spin correlators and coupling constants for a general class of quantum spin Hamiltonians bilinear in the spin operators has been recently established by Quintanilla, using an argument in the spirit of the Hohenberg-Kohn theorem in density functional theory. Quintanilla's theorem gives a firm theoretical foundation to quantum spin Hamiltonian learning using spin structure factors as input data. Here we extend the validity of the theorem in two directions. First, following the same approach as Mermin, the proof is extended to the case of finite-temperature spin structure factors, thus ensuring that the application of this theorem to experimental data is sound. Second, we note that this theorem applies to all types of Hamiltonians expressed as sums of bilinear operators, so that it can also relate the density-density correlators to the Coulomb matrix elements for interacting electrons in the lowest Landau level.

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Understanding the wonders and complexities of the microscopic world requires tackling the notoriously hard quantum many-body problem. Given the ubiquity of approximate methods in the state-of-the-art research on quantum many-body phenomena, the existence of general theorems [1–4] is essential for setting such approximations on firm theoretical ground.

The present Letter follows a recent work by Quintanilla [5], which establishes a theorem valid for a general class of bilinear quantum spin Hamiltonians,

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

where $\hat{S}_i^\alpha$ is the $\alpha = x, y, z$ component of a spin operator acting on site i in an arbitrary lattice, and $J_{i,j}^{\alpha,\beta}$ are the spin coupling constants. The general Hamiltonian stated in Eq. (1) encompasses most physically relevant types of interactions, notably Heisenberg [6], dipolar, Ising [7], Dzyaloshinskii-Moriya [8,9] and Kitaev [10] interactions. As a result, a wide class of canonical quantum spin models (e.g., Ising [7], Heisenberg [6], XXZ [11], Majumdar-Ghosh [12], Shastry-Sutherland [13], Haldane [14], Kitaev [10]) are particular cases of Eq. (1). It should be noted, however, that this class of Hamiltonians does not cover important quantum spin models such as the toric code [15] or the bilinear-biquadratic Heisenberg model (including the AKLT model [16]), which is known to describe some physical systems [17].

The theorem proven by Quintanilla [5] for this class of bilinear spin Hamiltonians asserts that there exists a one-to-one correspondence between the exchange constants $J_{i,j}^{\alpha,\beta}$ and the zero-temperature correlators

$$\rho_{i,j}^{\alpha,\beta}(T=0) = \langle \Phi_0 | \hat{S}_i^\alpha \hat{S}_j^\beta | \Phi_0 \rangle \tag{2}$$

for a physical system represented by the wave function $|\Phi_0\rangle$, which corresponds to the nondegenerate [18] ground state of a Hamiltonian of the form given in Eq. (1). The proof of Quintanilla's theorem runs parallel to that of the Hohenberg-Kohn theorem [1] for density functional theory. Interestingly, Mermin generalized [2] the Hohenberg-Kohn theorem to finite temperature, which motivates us to look for a finite-temperature generalization of Quintanilla's theorem as well.

In addition to the aforementioned theorem on the bijection between the exchange constants $J_{i,j}^{\alpha,\beta}$ and the zero-temperature spin-spin correlators $\rho_{i,j}^{\alpha,\beta}(T=0)$, Quintanilla also proved [5] a second theorem that establishes a one-to-one relation between $\rho_{i,j}^{\alpha,\beta}(T=0)$ and the ground state wave function $|\Phi_0\rangle$. This Letter will focus mainly on the first theorem, given its potential relevance within the context of the Hamiltonian learning problem for both the study of complex quantum condensed-matter systems and the verification of quantum technologies (cf. [19] and references therein). In any case, we will also briefly discuss the extension of this second theorem to finite temperature below.

Discussion. Before proceeding to the extension of Quintanilla's first theorem to the case of finite temperature, we begin by introducing some relevant concepts and notations. The thermal spin correlators at temperature T are defined as

$$\rho_{i,j}^{\alpha,\beta}(\hat{W}) := \mathrm{Tr}\big(\hat{W} \hat{S}_i^\alpha \hat{S}_j^\beta\big), \tag{3}$$

where $\hat{W} = \sum_n \frac{e^{-\beta E_n}}{Z} |\Phi_n\rangle\langle\Phi_n|$ is the density operator, $Z = \sum_n e^{-\beta E_n}$ is the partition function, $\{E_n\}$ and $\{|\Phi_n\rangle\}$ are the eigenenergies and eigenstates of a Hamiltonian $\hat{H}$ of the form

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given in Eq. (1), and $\beta^{-1} = k_B T$. Expanding Eq. (3) in the Hamiltonian eigenbasis gives

$$\rho_{i,j}^{\alpha,\beta}(\hat{W}) = \frac{1}{Z}\sum_n e^{-\beta E_n}\langle\Phi_n|\hat{S}_i^\alpha \hat{S}_j^\beta|\Phi_n\rangle. \tag{4}$$

Setting $T = 0$, or $\beta \to \infty$, in Eq. (4) results in Eq. (2), as expected. Spin-spin correlators can be measured experimentally using neutron diffraction [20,21].

In the following we show that the mapping between the coupling constants $J_{i,j}^{\alpha,\beta}$ and the *finite-temperature* correlators [cf. Eq. (4)] is bijective. We restrict ourselves to finite temperatures, since for $T \to \infty$ all but local $\langle\hat{S}_i^z \hat{S}_i^z\rangle$ correlators vanish (cf. Appendix).

The proof follows in a similar vein to the zero-temperature one by Quintanilla [5], but the Rayleigh-Ritz variational principle is replaced by the Gibbs-Bogoliubov inequality [2,22] for the Helmholtz free energy. Let a system described by a Hamiltonian $\hat{H}$ be in contact with a thermal bath at temperature T. The Helmholtz free energy of such system is

$$F(\hat{W}) = -k_B T \ln Z = \langle\hat{H}\rangle_{\hat{W}} - TS[\hat{W}], \tag{5}$$

where $\langle\hat{O}\rangle_{\hat{W}} = \mathrm{Tr}(\hat{W}\hat{O}) = \frac{1}{Z}\sum_n e^{-\beta E_n}\langle\Phi_n|\hat{O}|\Phi_n\rangle$ is the expectation value of some operator $\hat{O}$ at finite temperature and $S[\hat{W}] = -k_B \mathrm{Tr}(\hat{W}\ln\hat{W})$ is the von Neumann entropy. The Gibbs-Bogoliubov inequality sets an upper bound on the Helmholtz free energy,

$$F(\hat{W}) \leqslant \langle\hat{H}\rangle_{\hat{W}'} - TS[\hat{W}'], \tag{6}$$

for any positive semidefinite operator $\hat{W}'$ of appropriate dimensionality. The equality in Eq. (6) only occurs either when $\hat{W} = \hat{W}'$ or $T \to \infty$. A proof of the Gibbs-Bogoliubov inequality can be found in [2].

The proof of Quintanilla's first theorem at finite temperature proceeds by *reductio ad absurdum*. We consider two different Hamiltonians of the form given in Eq. (1), $\hat{H}$ and $\hat{H}'$. Their corresponding coupling constants, $J_{ij}^{\alpha,\beta}$ and $J_{ij}'^{\alpha,\beta}$, cannot therefore be all equal in pairs. Since the coupling constants determine the energies and eigenstates, they determine $\hat{W}$ and $\hat{W}'$ as well, the equilibrium density operators for $\hat{H}$ and $\hat{H}'$, respectively. We then assume that both $\hat{W}$ and $\hat{W}'$ are associated with the same finite-temperature spin-spin correlators,

$$\rho_{i,j}^{\alpha,\beta}[\hat{W}] = \rho_{i,j}^{\alpha,\beta}[\hat{W}'], \tag{7}$$

for all i, j, α, β. We can use the Gibbs-Bogoliubov inequality to write the following expression for the Helmoltz free energy of the unprimed system:

$$\begin{aligned} F(\hat{W}) &= \sum_{i,j,\alpha,\beta} J_{ij}^{\alpha,\beta}\rho_{i,j}^{\alpha,\beta}[\hat{W}] - TS[\hat{W}] \leqslant \sum_{i,j,\alpha,\beta} J_{ij}^{\alpha,\beta}\rho_{i,j}^{\alpha,\beta}[\hat{W}'] - TS[\hat{W}'] = \\ &= \sum_{i,j,\alpha,\beta}\left(J_{ij}^{\alpha,\beta} - J_{ij}'^{\alpha,\beta}\right)\rho_{i,j}^{\alpha,\beta}[\hat{W}'] + F(\hat{W}'). \end{aligned} \tag{8}$$

We can now exchange the roles of $\hat{W}$ and $\hat{W}'$ to obtain an identical expression for the primed system:

$$F[\hat{W}'] \leqslant \sum_{i,j,\alpha,\beta}\left(J_{ij}'^{\alpha,\beta} - J_{ij}^{\alpha,\beta}\right)\rho_{i,j}^{\alpha,\beta}[\hat{W}] + F(\hat{W}). \tag{9}$$

Summing Eqs. (8) and (9) yields

$$F[\hat{W}] + F[\hat{W}'] \leqslant \sum_{i,j,\alpha,\beta}\left(J_{ij}'^{\alpha,\beta} - J_{ij}^{\alpha,\beta}\right)\left(\rho_{i,j}^{\alpha,\beta}[\hat{W}] - \rho_{i,j}^{\alpha,\beta}[\hat{W}']\right) + F[\hat{W}] + F[\hat{W}']. \tag{10}$$

Using Eq. (7) turns Eq. (10) into $F[\hat{W}] + F[\hat{W}'] \leqslant F[\hat{W}] + F[\hat{W}']$. The equality holds only in the two trivial limits of infinite temperature or $\hat{H} = \hat{H}'$. For finite temperature and $\hat{H} \neq \hat{H}'$, we can replace the symbol $\leqslant$ by a strict inequality, thus arriving at a contradiction: $F[\hat{W}] + F[\hat{W}'] < F[\hat{W}] + F[\hat{W}']$. It follows, then, that the initial assumption stated in Eq. (7) must be false, in which case we can conclude that the *finite-temperature* correlators are single-valued functions $\rho_{ij}^{\alpha,\beta}[\hat{W}]$ of the equilibrium density operator $\hat{W}$, which is, in turn, uniquely determined by the coupling constants $\{J_{ij}^{\alpha,\beta}\}$ of the model, so that $\rho_{ij}^{\alpha,\beta}(J_{ij}^{\alpha,\beta})$ is injective.

Proving the injectivity of $\rho_{ij}^{\alpha,\beta}(J_{ij}^{\alpha,\beta})$ suffices to show it is bijective (i.e., a one-to-one mapping) since $\rho_{ij}^{\alpha,\beta}(J_{ij}^{\alpha,\beta})$ is surjective by construction, assuming, of course, that the physical system under study can be described by a Hamiltonian of the form given in Eq. (1). Indeed, given a model defined by a set of coupling parameters $\{J_{ij}^{\alpha,\beta}\}$, we can always determine, at least in principle, the respective equilibrium density operator $\hat{W}$, which can then be used to compute the finite-temperature spin-spin correlators $\{\rho_{ij}^{\alpha,\beta}\}$ per Eq. (4). This is entirely analogous to the trivial surjectivity of the mapping of the ground state wave functions onto the set of number densities in the proof of the Hohenberg-Kohn theorem [23]: every number density must be associated with a given wave function. Interestingly, the one-to-one relation between the ground state wave function and the external potential in spin-density-functional theory is not guaranteed to hold [24].

The final step amounts to recognizing that bijectivity is a sufficient condition for a function to be invertible. Hence, the coupling constants $\{J_{ij}^{\alpha,\beta}\}$ are themselves single-valued functions of the *finite-temperature* correlators $\{\rho_{ij}^{\alpha,\beta}\}$, which concludes the generalization of Quintanilla's first theorem to the case of finite temperature. Importantly, this result involves not only the ground state manifold (regardless of its degeneracy) but *excited states* as well.

As in the case of the Hohenberg-Kohn theorem, the present theorem does not give a systematic method to obtain the functional that relates the coupling constants $J_{ij}^{\alpha,\beta}$ to the correlators $\rho_{ij}^{\alpha,\beta}$. Hence, Quintanilla's first theorem does not produce a practical short-term advantage to tackle quantum spin Hamiltonians. Nevertheless, this theorem does provide a solid theoretical basis for a novel approach to the important problem of determining the parent Hamiltonian of experimental systems.

Artificial intelligence methods have been used to infer spin couplings out of experimentally determined spin correlators in spin-ice compounds [25]. The process includes the training of an artificial neural network (ANN) based on classical spin model simulations. In a similar vein, ANNs have been trained to infer spin couplings out of specific heat measurements [26]. We note that our finite-temperature theorem provides not only a firm foundation but also a practical advantage to infer spin couplings out of spin correlators, as it opens the possibility of training an ANN with simulations of quantum spin models. As noted by Yu *et al.* [26], this process is actually simplified at large temperatures. The computational resources needed to carry out exact diagonalizations of spin Hamiltonians scale exponentially with the system size. At high temperatures, however, the spatial range of spin correlators is expected to be shorter, which provides a natural cut-off for the size of the simulation cells [26]. We also note that the training could be supported by digital quantum simulations of quantum spin models [27] on noisy intermediate-scale quantum computers [28] using hybrid variational algorithms [29]. This approach may be used to accurately determine the spin Hamiltonian of Kitaev materials, such as $RuCl_3$ [30,31]. Likewise, it may assist in the characterization of state-of-the-art quantum technologies [19,32–35].

For the sake of completeness, we also discuss the extension of Quintanilla's second theorem to nonzero temperature. The original version at $T = 0$ states that the ground state wave function $|\Phi_0\rangle$ is uniquely determined by the spin-spin correlators $\rho_{i,j}^{\alpha,\beta}(T = 0)$. We note that this second theorem is not a mere corollary of the first, as the search for the ground state wave function, given the spin-spin correlators, is carried out in a Hilbert space encompassing all spin states of appropriate dimensionality, including those that are *not* eigenstates of a bilinear Hamiltonian of the form stated in Eq. (1).

At $T \neq 0$, the relevant physical description of the system is in terms of the density operator $\hat{W} = \sum_n \frac{e^{-\beta E_n}}{Z} |\Phi_n\rangle\langle\Phi_n|$. Following the argument of the proof at $T = 0$, we consider a given Hamiltonian of the form defined in Eq. (1), the equilibrium state of which is determined by the density operator $\hat{W}$ at some temperature T. By hypothesis, we assume there is some other density operator $\hat{W}' \neq \hat{W}$ such that both are associated with the same spin-spin correlators, as in Eq. (7). The Helmholtz free energy of the system considered can be expressed as

$$F[\hat{W}] = \sum_{i,j,\alpha,\beta} J_{ij}^{\alpha,\beta} \rho_{i,j}^{\alpha,\beta}[\hat{W}] - TS[\hat{W}] < \\ < F[\hat{W}'] = \sum_{i,j,\alpha,\beta} J_{ij}^{\alpha,\beta} \rho_{i,j}^{\alpha,\beta}[\hat{W}'] - TS[\hat{W}'], \qquad (11)$$

where the Gibbs-Bogoliubov inequality was used in the second step. As discussed above, a strict inequality can be assumed if one ignores the trivial case of $T = \infty$. Using Eq. (7) to simplify the inequality above gives $TS[\hat{W}] > TS[\hat{W}']$. Setting $T = 0$, we retrieve the result previously derived by Quintanilla [5], since we arrive at a contradiction $(0 > 0)$.

For $T > 0$, however, the entropic term does not vanish. Hence, a contradiction can be avoided if the von Neumann entropy of $\hat{W}'$ is lower than that of $\hat{W}$. The translation of Quintanilla's second theorem to finite temperature is therefore a weaker version of the zero-temperature counterpart: It merely asserts that, having obtained a density operator estimate $\hat{W}$ from a machine-learning model that fits the experimentally-obtained finite-temperature two-point correlators $\rho_{i,j}^{\alpha,\beta}$, there is no other density operator $\hat{W}'$ of *equal or greater entropy* that fits the data as well as $\hat{W}$. In other words, the system is *only* guaranteed not to be more disordered than the current prediction.

Returning to the first theorem, we note that, in all of the above we never make use of the fact that $\hat{S}_i^\alpha$ are spin operators. Hence, the theorem applies to any Hamiltonian that can be expressed as a bilinear sum of operators,

$$\hat{H} = \sum_{a,b} J_{a,b} \hat{O}_a \hat{O}_b, \qquad (12)$$

where $J_{a,b}$ describe couplings between operators $\hat{O}_a$ and $\hat{O}_b$, with a and b general labels. For example, Hamiltonian (12) includes the relevant case of interacting electrons that occupy a flat band or a single Landau level, with $\hat{O}$ being the electronic density operator and $J_{a,b}$ being the Coulomb interaction projected onto the lowest Landau level [36].

The first theorem stated and proven above can be rephrased as follows. The couplings $J_{a,b}$ are a single-valued functional of the thermal correlators:

$$\rho_{a,b} = \frac{1}{Z} \sum_n e^{-\beta E_n} \langle\Phi_n|\hat{O}_a \hat{O}_b|\Phi_n\rangle. \qquad (13)$$

Thus, the theorem establishes a one-to-one correspondence between the finite-temperature density-density correlations and the representation of the Coulomb matrix elements in the lowest Landau level. Experimentally, the scattering of charged particles is a natural probe of density-density correlation functions [37], but in practice it is problematic in the presence of strong magnetic fields. Still, density-density correlators can be probed in the case of atomic quantum gases, for which synthetic magnetic fields can be created [38]. We note that, most certainly, the functional relation between density-density correlators and Coulomb matrix elements will include an explicit dependence on the number of electrons in the system, as it is well known that the electronic properties of the two-dimensional electron gas in the lowest Landau level depend strongly on the filling factor [39].

The Hohenberg-Kohn theorem [1], and Mermin's finite-temperature extension [2], became extremely useful when approximate versions of the density functional, such as the Kohm-Sham local density approximation [40], were developed. We hope that this paper will inspire the quest for such approximate functionals within the context of quantum spin

Hamiltonian learning based on spin structure factors. We also note the existence of a theorem relating the ground state energy to the magnetization density in Heisenberg models [41]; its connection with our work remains to be explored.

Conclusion. We have demonstrated a theorem that establishes a one-to-one relation between interaction couplings and finite-temperature correlators in a general class of bilinear Hamiltonians. Our work generalizes a recent result of Quintanilla [5] in two ways. First, our theorem establishes the validity of Quintanilla's result for arbitrary temperatures. Second, we note that the theorem is applicable beyond the realm of spin systems. Our theorem puts the recent work that uses artificial intelligence to determine spin couplings [25] on firm theoretical footing and may provide a route to settle disputes about the nature of spin couplings in quantum materials, such as Kitaev materials, and to characterize state-of-the-art quantum technologies.

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APPENDIX: INFINITE-TEMPERATURE SPIN-SPIN CORRELATORS

The spin-spin correlators $\rho_{ij}^{\alpha\beta}(\hat{W})$ at a nonzero temperature T are given by Eq. (4). Setting $T\to\infty$, or $\beta\to 0$, gives $e^{-\beta E_n}=1$ for all eigenenergies $\{E_n\}$, in which case

$$\rho_{ij}^{\alpha\beta}(T\to\infty)=\frac{1}{Z}\sum_n\langle\Phi_n|\hat{S}_i^\alpha\hat{S}_j^\beta|\Phi_n\rangle\equiv\frac{1}{Z}\mathrm{Tr}\big(\hat{S}_i^\alpha\hat{S}_j^\beta\big).$$

Being a scalar, $\rho_{ij}^{\alpha\beta}(T\to\infty)$ is invariant under a change of basis. Since the density operator $\hat{W}$ now only contributes a constant prefactor $\frac{1}{Z}$, we can replace the Hamiltonian eigenbasis $\{|\Phi_n\rangle\}$ with the product basis $\{\bigotimes_i|S_i\rangle\}$, where we define the quantization axis such that $\hat{S}_i^z|S_i\rangle=S_i|S_i\rangle$ at every site i, with $S_i\in\{-S,-S+1,...,-1,0,1,...,S-1,S\}$ for a local spin-S. Computing the trace in this product basis gives

$$\rho_{ij}^{\alpha\beta}(T\to\infty)=\frac{\delta_{\alpha z}\delta_{\beta z}}{(2S+1)^2}\sum_{S_i,S_j=-S}^{S}\langle S_i|\hat{S}_i^\alpha|S_i\rangle\langle S_j|\hat{S}_j^\beta|S_j\rangle,$$

where the Kronecker deltas follow from the fact that the only spin component with nonzero entries along the diagonal is $\hat{S}^z$, so for any choice other than $(\alpha,\beta)=(z,z)$ the correlator vanishes. The prefactor results from the fact that the trivial sums over the remaining $N-2$ spins-S give a factor $(2S+1)^{N-2}$, which cancels with the partition function $Z=(2S+1)^N$.

Considering, for the moment, the nonlocal case $i\neq j$, we realize that for every configuration where $\langle S_i|\hat{S}_i^z|S_i\rangle\langle S_j|\hat{S}_j^z|S_j\rangle$ takes a value c, there is another one taking the symmetric value $-c$. In other words, any term $\langle S|\hat{S}_i^z|S\rangle\langle S'|\hat{S}_j^z|S'\rangle$ is canceled out by another term $\langle -S|\hat{S}_i^z|-S\rangle\langle S'|\hat{S}_j^z|S'\rangle$. Hence, all nonlocal spin-spin correlators vanish at infinite temperature:

$$\rho_{ij}^{\alpha\beta}(T\to\infty)=\frac{\delta_{ij}\delta_{\alpha z}\delta_{\beta z}}{2S+1}\sum_{S_i=-S}^{S}\langle S_i|(\hat{S}_i^z)^2|S_i\rangle.$$

The remaining sum can be computed explicitly: $\sum_{S_i=-S}^{S}S_i^2=\frac{S(S+1)(2S+1)}{3}$. Replacing in the expression above yields

$$\rho_{ij}^{\alpha\beta}(T\to\infty)=\delta_{ij}\delta_{\alpha z}\delta_{\beta z}\frac{S(S+1)}{3}.$$

Of course, this result is valid for any Hamiltonian, since neither the eigenspectrum nor the eigenstates appear in any step of this calculation at infinite temperature.

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