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We introduce a well-defined and unbiased measure of the strength of correlations in quantum many-particle systems which is based on the relative von Neumann entropy computed from the density operator of correlated and uncorrelated states. The usefulness of this general concept is demonstrated by quantifying correlations of interacting electrons in the Hubbard model and in a series of transition-metal oxides using dynamical mean-field theory.

Correlations in solids are the origin of many surprising phenomena such as Mott-insulating behavior and high-temperature superconductivity [1–3]. During the last few years the investigation of correlations between ultracold atoms in optical lattices has opened another fascinating field of research [4].

In many-body physics, correlations are conventionally defined as the effects which go beyond factorization approximations such as Hartree-Fock theory [5]. The actual strength of correlations in a given system is usually quantified by an interaction strength U relative to an energy unit such as the bandwidth W, or by comparing expectation values of particular operators, e.g., the interaction or the total energy [6–10]. However, there are many other quantities which can in principle be used to measure the correlation strength. Indeed "correlations" are by definition a relative concept since they always require the comparison with some reference system. Any approach which employs the expectation value of a particular set of operators for comparison will be biased. This raises a fundamental question: is there an objective way to quantify the correlations of a system, which even allows one to compare the correlation strength of different systems?

Maximal information about a general quantum state is provided by the corresponding density operator ("statistical operator") $\hat{\rho} = \sum_i p_i |\psi_i\rangle\langle\psi_i|$, where p_i is the probability for the quantum state $|\psi_i\rangle$ to be present in the mixture [11]. If there exists a basis in which $\hat{\rho}$ can be completely factorized ($\hat{\rho} \rightarrow \hat{\rho}^{PS}$, where PS refers to "product state"), the corresponding state is by definition uncorrelated [12]. To quantify the correlation strength of a quantum state one has to compare $\hat{\rho}$ with $\hat{\rho}^{PS}$ in a suitable way.

In this Letter we propose to quantify correlations within a statistical approach which is based on the concept of the von Neumann entropy [11,13]. We will show that the relative entropy [14] of a quantum state with respect to an uncorrelated product state provides a well-defined, unbiased, and useful measure of the correlation strength in that system. This concept not only allows one to quantify correlations, but even to compare the correlation strength of different quantum states. In quantum theory the von Neumann entropy is defined as $S(\hat{\rho}) = -\langle\ln\hat{\rho}\rangle_{\hat{\rho}} = -\text{Tr}(\hat{\rho}\ln\hat{\rho})$. It quantifies the degree to which a quantum system is in a mixed state. In this way one can also define a relative entropy [14]

$$\Delta S(\hat{\rho}\|\hat{\sigma}) \equiv -(\langle\ln\hat{\rho}\rangle_{\hat{\rho}} - \langle\ln\hat{\sigma}\rangle_{\hat{\rho}}), \tag{1}$$

provided the support [15] of $\hat{\rho}$ is contained in the support of $\hat{\sigma}$; otherwise $\Delta S(\hat{\rho}\|\hat{\sigma})$ is infinite. By definition the relative entropy defined in this way is positive, monotonic, additive, and convex in its arguments [16]. The relative entropy measures "how different" two quantum states are [14,16–19]. Therefore we propose to quantify correlations by the relative entropy $\Delta S(\hat{\rho}\|\hat{\rho}^{PS})$ between the statistical operator $\hat{\rho}$ of a system and an uncorrelated product state with $\hat{\rho}^{PS}$ for the same system [16,20]. Since this approach uses a statistical operator it accounts for correlations represented by all possible correlation functions generated by $\hat{\rho}$. In the following we will make use of a reduced statistical operator which restricts the set of assessable correlation functions.

The general approach works as follows [22–26]. Let $|\Psi\rangle$ be any state in the Hilbert space of the system. We decompose the system into two subsystems yielding two Hilbert subspaces $A = \{|a\rangle\}$ and $B = \{|b\rangle\}$, where $|a\rangle$ and $|b\rangle$ are basis vectors of these subspaces. Then one may write $|\Psi\rangle = \sum_{ab}\Psi_{a,b}|a\rangle|b\rangle$ with amplitudes $\Psi_{a,b}$. We trace out the degrees of freedom of the B subsystem. The reduced statistical operator is given by $\hat{\rho}_A^\Psi = \text{Tr}_B|\Psi\rangle\langle\Psi| = \sum_{a_1,a_2}|a_1\rangle\rho_{a_1,a_2}^\Psi\langle a_2|$, with the matrix elements $\rho_{a_1,a_2}^\Psi = \sum_b\Psi_{a_1,b}\Psi_{a_2,b}^*$. By defining diagonal operators, i.e., projectors, $\hat{P}_a = |a\rangle\langle a|$, and off-diagonal operators

$\hat{T}_{a_1 a_2} = |a_1\rangle\langle a_2|$, respectively, one can determine $\rho^{\Psi}_{a_1, a_2} = \langle\Psi|\hat{T}_{a_1, a_2}|\Psi\rangle^{\dagger}$ explicitly.

In the following, we will demonstrate the usefulness of this general concept by applying it to interacting lattice electrons described by the Hubbard model, i.e., a generic many-electron model with purely local interaction. In its simplest version it has the form

$$\hat{H} = \sum_{ij} t_{ij}\hat{c}^{\dagger}_{i\sigma}\hat{c}_{j\sigma} + U\sum_i \hat{n}_{i\uparrow}\hat{n}_{i\downarrow}, \tag{2}$$

where $\hat{c}^{\dagger}_{i\sigma}$ ($\hat{c}_{i\sigma}$) are creation (annihilation) operators for fermions of spin $\sigma = \uparrow$ ($\downarrow$) at the lattice site i, t_{ij} are hopping amplitudes between different sites, and U is the local interaction energy. The Hubbard model cannot be solved exactly. As an approximation we therefore employ dynamical mean-field theory (DMFT), which becomes exact in the limit of infinite spatial dimensions ($d \to \infty$) [27] and is known to provide a reliable local, but fully dynamical, description of correlated electron systems in $d = 3$ [28,29]. By using DMFT to compute the relative von Neumann entropy only the most important, namely, local, correlations of the exact solution are taken into account. In DMFT the subspace A is chosen to correspond to a site i with the four state vectors $|a\rangle = \{|0\rangle, |\uparrow\rangle, |\downarrow\rangle, |2\rangle\}$ which characterize the local occupation of the site. By computing the statistical operator within DMFT we determine the eigenvalues $p_a = \langle\Psi|\hat{P}_a|\Psi\rangle$ of the reduced statistical operator $\hat{\rho}_A$. We then employ an explicit product state $|PS\rangle$ to calculate the uncorrelated reduced statistical operator $\hat{\rho}^{PS}_A$ and its eigenvalues p^{PS}_a. In this way the relative entropy (1) of the two states can be determined. In the following, the subspace index $A \equiv i$ will be omitted. In the absence of off-diagonal long-range order ($\langle\Psi|\hat{c}_{\sigma}|\Psi\rangle = 0$, $\langle\Psi|\hat{c}_{\sigma}\hat{c}_{-\sigma}|\Psi\rangle = 0$) the reduced, i.e., local, statistical operator is diagonal [16,22–26]: $\hat{\rho} = p_0|0\rangle\langle 0| + \sum_{\sigma} p_{\sigma}|\sigma\rangle\langle\sigma| + p_2|2\rangle\langle 2|$. The diagonal matrix elements p_a are expressed by the particle number density n, magnetization density m, and double occupation per site d, i.e., $p_0 = 1 - n + d$, $p_{\sigma} = (n + \sigma m)/2 + d$, and $p_2 = d$. Because of its diagonal form the local von Neumann entropy is given by $S(\hat{\rho}) = -\sum_a p_a \log p_a$ with $a = 0, \uparrow, \downarrow, 2$. Then the local relative entropy becomes

$$\Delta S(\hat{\rho}\|\hat{\rho}^{PS}) = -\sum_a p_a(\ln p_a - \ln p^{PS}_a), \tag{3}$$

where p^{PS}_a is the corresponding expectation value within the factorization approximation corresponding to $|PS\rangle$. We note that the reversed relative entropy $\Delta S(\hat{\rho}^{PS}\|\hat{\rho})$ is also defined. The statistical operator is no longer diagonal if, for example, the local interaction includes nondensity type terms, or if superconducting phases are considered. In this case the statistical operator needs to be diagonalized to determine its logarithm.

To compute the parameters n, m, and d the DMFT self-consistency equations are solved numerically by using the numerical renormalization group method at zero temperature [30]. We assume hopping $t_{ij} \equiv t$ between nearest neighbor sites i, j on a cubic lattice and use the corresponding density of states for the DMFT calculations. The bandwidth $W = 6t = 1$ sets the energy unit. We fix the particle number density at $n = 1$. In the following both paramagnetic and antiferromagnetic ground states are considered.

(i) *Paramagnetic ground state.*—We analyze the correlated paramagnetic (PM) ground state $|PM\rangle$ with respect to the following two reference states: (i) the product state in k space $|PS\rangle \equiv |\text{free}\rangle$ corresponding to noninteracting ($U = 0$) electrons, where $d = (n^2 - m^2)/4$, and (ii) the product state in position space, i.e., the local moment state $|PS\rangle \equiv |LM\rangle$ where $d = 0$. In the PM phase $m = 0$ implying that $S(\hat{\rho}) = -2[d \ln d + (\frac{1}{2} + d)\ln(\frac{1}{2} + d)]$ is determined solely by the dependence of the double occupation d on U. In the upper panel of Fig. 1 the local entropy is shown as a function of U, and d is shown in the inset. Both are monotonically decreasing functions of U: the local entropy decreases from $\ln 4$ at $U = 0$ to $\ln 2$ at $U = \infty$ and the double occupation from $1/4$ to 0. At half-filling a Mott-Hubbard metal insulator transition (MIT) takes place at a critical interaction $U_c \approx 1.225$, at which a correlation gap opens in the one-particle spectral function. At the MIT the double occupation, and therefore the local entropy, displays a kink. The free state $|\text{free}\rangle$ and the local moment state $|LM\rangle$ have constant local entropies $S_{\text{free}} = \ln 4$ and $S_{\text{LM}} = \ln 2$, respectively, which reflect the degeneracy of those uncorrelated product states.
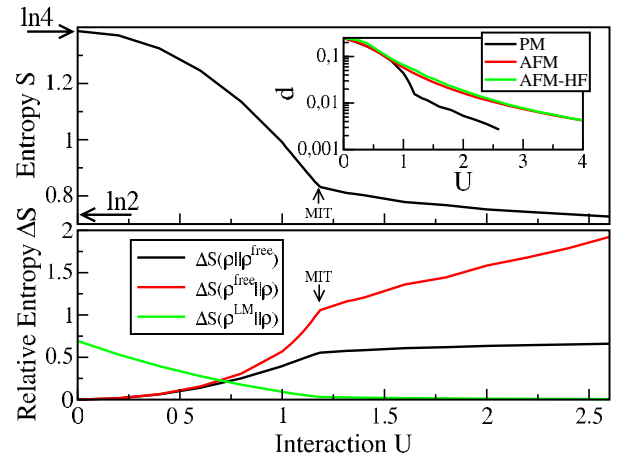

FIG. 1 (color online). Upper panel: Local von Neumann entropy of the correlated PM ground state as a function of the interaction U. Inset: double occupation in the PM (black curve) and the AFM (red curve) ground state compared to the Hartree-Fock result (green curve) as a function of U. Lower panel: local relative entropies (PM phase) between the correlated ground state and the noninteracting ($U = 0$) and local moment ($U = \infty$) ground states, respectively, as functions of U.

The correlated PM state |PM⟩ can be distinguished from the product states |free⟩ and |LM⟩, respectively, by means of four different relative entropies: $\Delta S(\hat{\rho}\|\hat{\rho}^{\text{free}})$, $\Delta S(\hat{\rho}\|\hat{\rho}^{\text{LM}})$, $\Delta S(\hat{\rho}^{\text{free}}\|\hat{\rho})$, and $\Delta S(\hat{\rho}^{\text{LM}}\|\hat{\rho})$, which are obtained from the three statistical operators $\hat{\rho}$, $\hat{\rho}^{\text{free}}$, and $\hat{\rho}^{\text{LM}}$ using Eq. (3). The relative entropies are functions of U as is shown in the lower panel of Fig. 1. The larger their value, the more distinguishable the two corresponding states are. In the Mott phase the relative entropy $\Delta S(\hat{\rho}\|\hat{\rho}^{\text{free}})$ increases much more slowly with the interaction strength U than in the metallic phase. This means that in a Mott insulator the correlation strength increases more slowly with U than in a correlated metal. We note that $\Delta S(\hat{\rho}\|\hat{\rho}^{\text{LM}}) = \infty$ for $U < \infty$ because in the local moment state double occupation is completely excluded ($p_2^{\text{LM}} = 0$), i.e., |PM⟩ and |LM⟩ can be perfectly distinguished by measuring the respective double occupation of the two states. By contrast, the reverse relative entropy $\Delta S(\hat{\rho}^{\text{LM}}\|\hat{\rho})$ remains finite although the two states are perfectly distinguishable. This shows that $\Delta S(\hat{\rho}\|\hat{\rho}^{\text{LM}})$ rather than the reverse expression is the more appropriate measure of the correlations.

(ii) *Antiferromagnetic case.*—Next we compare the correlated antiferromagnetic (AFM) ground state |AF⟩ with the product states represented by the Slater state |Slat⟩ (a spin-density product state in k space) and the Heisenberg state |Heis⟩ (a perfect Néel-type product state in position space), respectively. The staggered magnetization m_{st} of the Slater state is determined within Hartree-Fock, whence $d_{\text{Slat}} = [(n_{\text{Slat}})^2 - (m_{\text{st,Slat}})^2]/4$. In the Heisenberg state $m_{\text{st,Heis}} = 1$ and $d_{\text{Heis}} = 0$. As shown in the upper panel in Fig. 2 the local entropies $S(\hat{\rho})$ and $S(\hat{\rho}^{\text{Slat}})$ are almost identical. A similar agreement is found when the local entropies are plotted as functions of the order parameter m_{st} instead of the interaction U. Both local entropies decrease from ln4 at $U = 0$ to zero at $U = \infty$. This is

due to the presence of long-range order with a magnetization that fully saturates. It means that at $U = \infty$ the spin degeneracy is completely lifted. We observe that $S(\hat{\rho}) > S(\hat{\rho}^{\text{Slat}})$ because $m < m_{\text{Slat}}$ and $d < d_{\text{Slat}}$ due to quantum fluctuations, cf. inset of Fig. 1. The Slater (weak coupling) and the Heisenberg (strong coupling) limits are smoothly connected [31].

The correlated AFM state |AF⟩ can be distinguished from the product states |Slat⟩ and |Heis⟩, respectively, by the four relative entropies $\Delta S(\hat{\rho}\|\hat{\rho}^{\text{Slat}})$, $\Delta S(\hat{\rho}\|\hat{\rho}^{\text{Heis}})$, $\Delta S(\hat{\rho}^{\text{Slat}}\|\hat{\rho})$, and $\Delta S(\hat{\rho}^{\text{Heis}}\|\hat{\rho})$ which are obtained from the three statistical operators $\hat{\rho}$, $\hat{\rho}^{\text{Slat}}$, and $\hat{\rho}^{\text{Heis}}$. The relative entropies $\Delta S(\hat{\rho}\|\hat{\rho}^{\text{Slat}})$ and $\Delta S(\hat{\rho}^{\text{Slat}}\|\hat{\rho})$ are more than an order of magnitude smaller than in the PM case, as is seen in the inset to the upper panel of Fig. 2. As before the relative entropy $\Delta S(\hat{\rho}\|\hat{\rho}^{\text{Heis}}) = \infty$ because $d_{\text{Heis}} = 0$, implying $p_2^{\text{Heis}} = 0$ for any U. By contrast, $\Delta S(\hat{\rho}^{\text{Heis}}\|\hat{\rho})$ is finite and approaches zero at large U. Correlationwise the Heisenberg product state and the correlated AFM state are therefore very similar at large U, and cannot be easily distinguished. This means that, in contrast to the PM state, the AFM correlated state is locally only weakly correlated.

In a further application, we employ the relative entropy to quantify correlations in real materials. As an example, we select a series of transition-metal (TM) monoxides MnO, FeO, CoO, and NiO. The results are obtained within the LDA + DMFT approach [32–34], which combines the local density approximation (LDA) to density functional theory with the DMFT [16]. We take the states obtained from the LDA |LDA⟩ as the uncorrelated states. In Fig. 3 we show the local entropies of the series of TM monoxides. The local entropies computed by LDA + DMFT describe the corresponding ionic ground states, which correspond in all four cases to high-spin configurations. The orbital
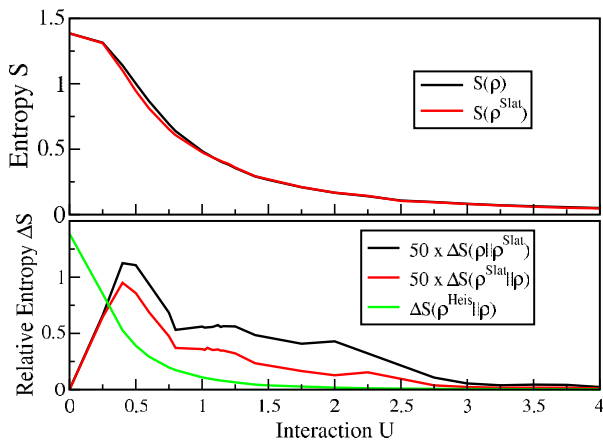

FIG. 2 (color online). Upper panel: local von Neumann entropy S of the correlated AFM and the Slater ground states as a function of U. Lower panel: local relative entropies ΔS between correlated AFM, Slater, and Heisenberg ground states. $\Delta S(\hat{\rho}\|\hat{\rho}^{\text{Slat}})$ and $\Delta S(\hat{\rho}^{\text{Slat}}\|\hat{\rho})$ are magnified by a factor of 50.
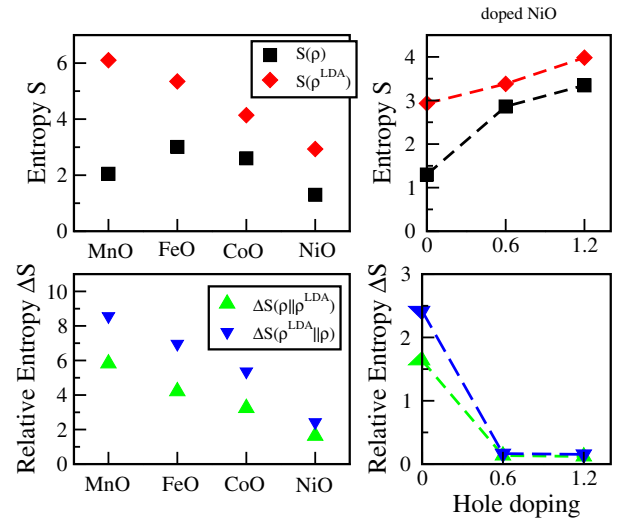

FIG. 3 (color online). Left panels: local von Neumann (top) and relative (bottom) entropies for MnO, FeO, CoO, and NiO series. Right panels: local von Neumann (top) and relative (bottom) entropies for hole doped NiO; see text.

degrees of freedom imply that NiO and MnO have a four-fold degeneracy, while FeO and CoO have a sixfold degeneracy [35]. The trend in the LDA entropy reflects the decreasing number of possibilities to distribute 5 (MnO), 6 (FeO), 7 (CoO), and 8 (NiO) electrons among 10 orbitals, which are modified by the crystal-field splitting and deviate from integer occupation. The dominant effect of the correlations is to reduce the large number of local many-body ground states available in the $|\text{LDA}\rangle$ solution to only a few in the $|\text{DMFT}\rangle$ solution. We conclude that MnO, with a d^5 configuration, is more correlated than NiO which has a d^8 configuration, because in a d^5 configuration the interaction modifies the local occupation matrix more significantly.

Finally, we investigate the effect of hole doping on NiO [34]. Reducing the $3d$ occupation towards half-filling increases the number of local states with non-negligible contribution to the ground state and leads to an increase of the local entropy with hole doping in the uncorrelated system. In the correlated system the local density matrix changes substantially. While in stoichiometric NiO the d^8 ($t_{2g}^6 e_g^2$) states dominate the local density matrix, hole doping leads to a progressive population of d^7 and d^9 ($t_{2g}^6 e_g^{1,3}$), respectively. The increase of the number of states which effectively contribute to the ground state is reflected in the increase of the local entropy. This leads to a lowering of the relative entropy with the number of holes and to a substantial decrease of correlations.

In conclusion, we employed the relative von Neumann entropy between correlated and factorized (uncorrelated) statistical operators to quantify the correlation strength in quantum many-body systems. We demonstrated the usefulness of this approach by computing the degree of correlation in the Hubbard model and in a series of transition monoxides using DMFT and LDA + DMFT, respectively. It will be interesting to apply this general concept to investigate correlations beyond a single-site approximation, in which case not only dynamical local, but also spatial correlations are included.

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