Long-range orbital order in a degenerate-orbital Hubbard model: absence in low-dimensions

Canio Noce
Laboratorio Regionale SuperMat, INFM-CNR, Salerno, Dipartimento di Fisica ‘E R Caianiello’, Università di Salerno, I-84081 Baronissi (Salerno), Italy
E-mail: canio@sa.infn.it.

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Abstract. We investigate the possibility of long range orbital order in low-dimensional systems. The study is performed within a two-orbital Hubbard model by means of the Bogoliubov’s inequality. We show that, in one- and two-dimensions, and when a narrow conduction band is concerned, this model does not exhibit orbital order at any filling and any finite temperature.

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1. Introduction

In recent years, materials in which the interplay between charge, spin and orbital degrees of freedom plays an important role have attracted a lot of interest both from theoretical and experimental perspectives [1]. An important characteristic of these materials is the existence of several quantum states competing among them. As a typical example, we may refer to the complicated phase diagrams that transition metal oxides (TMO) exhibit [2]. These systems are not as simple as standard metals: the many active degrees of freedom interact in a complex and synergic way so leading to an intrinsic complexity. The minimal model that can be used to describe the physics of TMO is certainly the Hubbard model which, in its simplest form, contains the local Coulomb interaction between the electrons in a conduction band formed by a single orbital only. The investigation of such a single-band model might be justified in some cases, in particular, as an effective model for low-energy properties. Nevertheless, it is widely accepted that in several 3d and 4d TMO [3], as well as in alkali-doped fullerides [4], a consistent description of the experimental observations requires the use of more realistic models where several interactions, between spins, charges and orbitals, are simultaneously active. When the orbital degree of freedom is taken into account, many interesting phenomena come in, allowing for the explanation of remarkable properties like colossal magnetoresistance [5], metal–insulator transition [6], and for properties involving orbital order. In particular, canonical examples of orbitally ordered compounds include YTiO$_3$ [7], YVO$_3$ [8], KCuF$_3$ [9] and perovskite manganites [10], while, recently, evidence of orbital order has been reported in the lanthanum ruthenate La$_4$Ru$_2$O$_{10}$ [11].

From the theoretical point of view, many approaches have been so far proposed to describe the effect of strong Coulomb interaction in systems with two-orbital degeneracy. Among them we quote the slave-boson methods [12], the variational method [13], and the limit of high spatial dimensions [14]. Nevertheless, theoretical advances in the comprehension of these systems are mainly due to the application of dynamical mean-field theory which has led to an increased understanding of correlation effects associated with the Mott metal–insulator transition [15]. Indeed, due to the presence of orbital degrees of freedom, the Mott physics contains extra elements of unconventional character. Recent results have indicated the possibility of orbital selective Mott transitions showing that separate Mott transitions occur at different Coulomb strengths, eventually merging into a single critical point for special conditions [16]. We would like to point out that the phenomenology of the Mott physics in this type of system has been intensively investigated [17, 18], and recent remarkable achievements show unambiguously that two successive and distinct Mott transitions are exhibited by systems described by the two-orbital Hubbard model [19, 20], further supporting the relevance of the orbital degeneracy.

On the other hand, referring to exact results, they are rather rare at present. Nevertheless, it has been recently shown that symmetry features of the ground state at half filling can be obtained in a large range of the parameter space, combining the property of spin reflection positivity and the use of special unitary transformations [21]. In particular, it has been possible to extract relevant symmetry characteristics related to the orbital, spin and $\eta$ pairing pseudospin operator, and, at half-filling and for different regions of the parameter space, the spin, orbital and $\eta$ pairing pseudospin quantum numbers of the ground state have been determined. Also, making use of these results, it has been rigorously shown that, at half-filling, the charged gaps are always larger than the spin-excitation gaps and properly defined orbital gaps [22]. For completeness, we also mention the $SU(4)$ theory presented for the one-dimensional model case [23].

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Here, our aim is to rigorously show that the two-orbital generalized Hubbard model with narrow bands does not exhibit orbital order. This result is based on the application of Bogoliubov’s inequality \[24\] and it holds true for any choice of the microscopic parameters of the model Hamiltonian and for any electron filling. We notice that this approach has been successfully, and routinely applied to many different strongly correlated electron models, usually ruling out the magnetic and the superconducting long range order at any finite temperature (for a review see \[25\]).

The paper is organized as follows: in the next section the model Hamiltonian is introduced and its symmetry properties are summarized. In section 3 the Bogoliubov’s inequality is applied to the model and the results are discussed; the last section is devoted to the conclusions.

2. The model and symmetry properties

The explicit form of the Hamiltonian for the two-orbital degenerate Hubbard model we refer to can be written as:

\[ H = H_t + H_{\text{int}}, \]  

where

\[ H_t = \sum_{\langle ij \rangle, \lambda, \sigma} t(R_i - R_j) d_{i, \lambda \sigma}^\dagger d_{j, \lambda \sigma}, \]  

\[ H_{\text{int}} = U_0 \sum_{i, \lambda} n_{i, \lambda \uparrow} n_{i, \lambda \downarrow} + U \sum_{i, \sigma} n_{i, \sigma} n_{i, \bar{\sigma}} + (U - J) \sum_{i, \sigma} n_{i, \sigma} n_{i, \bar{\sigma}} - J \sum_{i, \sigma} d_{i, \sigma}^\dagger d_{i, \bar{\sigma}} d_{i, \sigma} d_{i, \bar{\sigma}}. \]  

Here \( d_{i, \lambda \sigma}^\dagger \) is the creation operator for correlated electrons with spin \( \sigma \) at site \( i \) on orbital \( \lambda \) (\( \lambda = 1, 2 \)), and \( n_{i, \lambda \sigma} \) is the number operator for the electrons on the site \( i \) and orbital \( \lambda \). The overlap integrals \( t(R_i - R_j) \) are required to satisfy the parity symmetry \( t(R_i - R_j) = t(R_j - R_i) \) and they are assumed to be nonzero only for short ranged overlapping, i.e. we are considering narrow band systems, and we also suppose that \( \lambda \) orbitals are not mixed by the hopping. \( U_0, U \) and \( J \) stand for the intra-orbital, inter-orbital Coulomb and Hund’s rule exchange interaction, respectively. Finally, we have used the simplified notation \( \bar{\sigma} = -\sigma \).

The results presented below are valid for any choice of the parameters in the Hamiltonian. Nevertheless, since the two orbitals are equivalent, they can be interchanged by a properly chosen canonical transformation. Thus, we impose an additional condition on the set of the parameters, that is \( U_0 = U + 2J \) \[26\]. Hereafter, we will use this relation together with the condition that \( U \) and \( J \) are positive \[27\]. Hence \( H_{\text{int}} \) becomes,

\[ H_{\text{int}} = (U + 2J) \sum_{i, \lambda} n_{i, \lambda \uparrow} n_{i, \lambda \downarrow} + U \sum_{i, \sigma} n_{i, \sigma} n_{i, \bar{\sigma}} + (U - J) \sum_{i, \sigma} n_{i, \sigma} n_{i, \bar{\sigma}} - J \sum_{i, \sigma} d_{i, \sigma}^\dagger d_{i, \bar{\sigma}} d_{i, \sigma} d_{i, \bar{\sigma}}. \]  

Now, let us introduce the following operators \[21\]:

\[ S = \frac{1}{2} \sum_{i, \sigma, \sigma'} d_{i, \lambda \sigma}^\dagger (\sigma)_{\sigma' \sigma'} d_{i, \lambda \sigma'}, \]  

\[ T = \frac{1}{2} \sum_{i, \lambda, \lambda'} d_{i, \lambda \sigma}^\dagger (\sigma)_{\lambda \lambda'} d_{i, \lambda \sigma}, \]  

\[ \text{New Journal of Physics 9 (2007) 238 (http://www.njp.org/)} \]
\[ \eta = \frac{1}{2} \sum_{i, \sigma, \sigma'} \epsilon(i) d_{i \lambda \sigma}^{\dagger} (\sigma) \sigma' d_{i \lambda \sigma'}^{\dagger}, \]  

(7)

where \( \sigma \) are the Pauli matrices and \( \epsilon(i) = \pm 1 \) depending to which of the two subparts of the bipartite lattice the site \( i \) belongs.

These operators refer to the usual total spin operator \( S \), the orbital operator \( T \) and the so-called pairing operator \( \eta \) [28]. As for \( S \), \( T \) and \( \eta \) generate other SU(2) algebras. Furthermore, the total spin operator \( S \) commutes with the Hamiltonian \( [S, H] = 0 \) and it is a good quantum number. Another symmetry is the one related to the orbital degree of freedom as defined by \( T \); the square of the total orbital operator \( T \) and its third component \( T_z \) commute with the Hamiltonian \( H \), and the same happens for the square of the total \( \eta \) pseudospin operator and its third component \( \eta_z \). We point out that the algebras generated by these operators are not independent being related to each other by means of extended hole–particle and orbital type transformations [22].

In closing this section, we would like to discuss the properties of the orbital operator \( T \). We point out that the two possible choices of the orbitals are represented by \( T \), whose \( z \) component \( T_z = 1/2 \) when the orbital \( \lambda = 1 \) is occupied, and \( T_z = -1/2 \) when the other orbital \( (\lambda = 2) \) is occupied. Moreover, to address the issue quoted in the introduction section, we notice that when a long range orbital order exists, we have \( \langle T_{i z} \rangle \neq 0 \) [1].

Furthermore, we would like to emphasize that the Hamiltonian here adopted exhibits the conservation of the orbital quantum numbers and this condition would be of great help for the exact derivation presented below. Nevertheless, we want to point out that in some \( t_{2g} \) real systems like \( \text{LaTiO}_3 \) or \( \text{LaVO}_3 \), and \( e_g \) compounds, such as for instance \( \text{KCuF}_3 \), the theoretical description usually stems from spin–orbital model Hamiltonians with Coulomb interactions that prevent the conservation of \( T \) and the total third component operator \( T_z \) [29]. In addition, when other \( e_g \) systems like ferromagnetic manganites are considered, the presence of the off-diagonal hopping in the Hamiltonian gives rise to the absence of the \( SU(2) \) orbital invariance in the orbital space [30].

3. Bogoliubov’s inequality and orbital order

Let us discuss now how Bogoliubov’s inequality applies to this model Hamiltonian [25], confining the analysis to the orbital order.

When two quantum-mechanical operators \( A \) and \( B \) are considered, the Bogoliubov’s inequality reads as:

\[ \left| \langle [B^\dagger, A^\dagger] \rangle \right|^2 \leq \frac{\beta}{2} \left| \langle [A, A^\dagger] \rangle \right| \left| \langle [B^\dagger, [H, B]] \rangle \right|. \]

(8)

Here \( \langle \ldots \rangle \) stands for the thermal average over the grand canonical ensemble, \( \langle \ldots \rangle \) denotes the usual anticommutator and \( \beta = 1/k_B T \). To look for spontaneous charge order at finite temperature, following the Bogoliubov procedure, one first removes the degeneracy of the microscopic model Hamiltonian and then studies the expectation values involved in the inequality. The removal of the degeneracy is usually done by introducing a symmetry-breaking term into the Hamiltonian. To this end, we add to the Hamiltonian \( H \) the following symmetry breaking term relevant for the order under study:

\[ H_\omega = -\gamma \sum_i T_{i z} \exp(-i p R_x), \]

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where $\gamma$ is the amplitude of the symmetry-breaking term. This term simulates the interaction between a space-dependent static field $\gamma \exp(-ipR_z)$ along the $z$-axis and the orbital operator.

An adequate choice of the operators $A$ and $B$ in the Bogoliubov’s inequality has to be made in order to single-out the suitable order parameter. Therefore, we choose $A = T^-(k + p)$ and $B = T^+(k)$. Moreover, we notice that the Fourier transforms of the orbital operator components satisfy the commutation relations

$$
[T^+(k), T^-(k')] = \mp T^+(k + k'), \quad [T^+(k), T^-(k')] = 2T^z(k + k');
$$

one also has $[T^+(k)]^\dagger = T^-(k)$.

Let us search now for nontrivial upper bounds in the inequality equation (8).

After some straightforward, but lengthy algebra, we deduce the following inequality for the commutator containing the Hamiltonian:

$$
\langle [B^\dagger, [H, B]] \rangle \leq \left| \sum_{ij\sigma} t(R_i - R_j) \left\{ 1 - \exp[i(k_i - k_j)] \right\} \left( \delta_{ij\sigma} \delta_{j1\sigma} - \delta_{j2\sigma} \delta_{21\sigma} \right) + 2\gamma T^z(-p) \right|.
$$

Using the the following inequalities: (i) $|\sum_{k\sigma} (n_{k\sigma} - n_{k2\sigma})| \leq N_d$, $N_d$ being the total number of electrons in the two orbitals; (ii) $|1 - \cos x| < x^2/2$; (iii) the triangle inequality, together with the translational invariance of the model, one can write

$$
\langle [B^\dagger, [H, B]] \rangle \leq \sum_i |t(R_i)| \frac{R_i^2 k^2}{2} N_d + 2\gamma |\langle T^z(-p) \rangle|.
$$

Here, the quantity $\Delta \equiv \sum_i |t(R_i)| R_i^2 / 2$ is well defined since $t(R_i)$, the matrix elements of the overlap integral between Wannier functions, decay rapidly with distance. Indeed, when $t(R_i - R_j) = \gamma t$ for $i$ and $j$ nearest-neighbours, $\Delta = t z$, $z$ being the coordination number while in the case of next nearest-neighbours hopping integral $t'$, $\Delta = t z + t' z'$, where $l = 4(2)$ in one-(two)-dimension(s) and $z'$ is the number of next nearest-neighbour sites.

The proof of the absence of orbital order is accomplished if we show that, in the thermodynamic limit, the appropriate order parameter vanishes.

Summing both sides of equation (8) over $k$ and considering that

$$
\sum_k \langle [T^-(-k + p), T^+(k + p)] \rangle = N \sum_i \langle [T_i^-, T_i^+] \rangle \leq 4NN_d,
$$

we finally have

$$
\omega(p) = \frac{\beta NN_d}{2} \left[ \sum_k \frac{1}{\Delta N d k^2 + 2\gamma N \omega(p)} \right]^{-1},
$$

where we have introduced the following quantity $\omega(p) = |\langle T^z(-p) \rangle|/N$ which plays the role of the orbital order parameter.

When we replace the sum by the integral over the first Brillouin zone in the above inequality and take the thermodynamic limit, we obtain, for small $\gamma$,

$$
\omega(p) \leq C_1 \beta |\gamma|,
$$

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in one-dimension, and
\[
\omega(p) \leq C_2 \sqrt{\beta} \frac{1}{\sqrt{\ln|\gamma|}},
\]
(13)
in two-dimensions. Here, \(C_1, C_2\) are two constants independent of the temperature and on the symmetry breaking amplitude \(\gamma\).

From these inequalities, it follows that the orbital order parameter \(\omega(p)\) goes to zero when the amplitude of the symmetry breaking field \(\gamma\) vanishes, both in one-and-two-dimensions. As \(\omega(p)\) is proportional to the difference between the total number of electrons in orbitals 1 and 2, we conclude that there is no orbital order in the occupation of these two orbitals.

4. Conclusions

In this paper, we have rigorously shown that, applying Bogoliubov’s inequality, the two-orbital Hubbard model with narrow bands does not exhibit orbital order at any nonzero temperature in low-dimensional cases. This result is valid for any value of the Coulomb interactions and for any electron filling.

Nevertheless, we want to note that, the non-existence of an ordered phase at finite temperature does not necessarily mean that there cannot be a phase transition at finite temperature. We may indeed envisage a Kosterlitz–Thouless type transition to a phase with an algebraic fall-off of correlations \([31]\).

Moreover, the above mentioned considerations, as well as the rigorous proof presented in section 3, does not apply at \(T = 0\) implying that the ground state may be ordered. Indeed, in some situations an ordered phase is also possible at zero temperature and in low-dimensional cases, usually in two-dimensions. In this latter case, quantum fluctuations oppose but do not prevent the appearance of an ordered phase. In contrast, for one-dimensional systems, quantum fluctuations are so strong that they prevent long-range order in the ground state, as is well known for the Heisenberg \([32]\), the Hubbard \([33]\) and the Anderson models \([34]\).

We want to stress that we have confined the analysis of the present paper to the study of the absence of orbital order, since this order has never been studied before by means of Bogoliubov’s inequality. Of course, the study of magnetic and superconducting long range order could be easily done following the procedure here outlined. In this respect, we note that such an analysis has been recently performed in a multi-orbital model suitably chosen to study orbitally degenerate systems. Specifically, starting from the Kugel–Khomskii Hamiltonian, adopted to investigate the properties of a threefold degenerate \(t_{2g}\) orbitals for cubic titanates \([35]\), the application of Bogoliubov’s inequality has shown that a simplified version of the above mentioned model, i.e. in the absence of Hund’s exchange, does not support spontaneous long-range spin order at any nonzero temperature and for any value of the model parameters \([36]\).

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Corrigendum added 16 November 2007

On page 3 of this paper, the sentence:

‘Thus, we impose an additional condition on the set of the parameters, that is $U_0 = U + 2J$ [26].’

should be replaced by the following:

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and reference [26] should be replaced by:

[26] Oleš A M 1981 Phys. Rev. B 23 271