Charge transfer and the statistics of holons in a periodical lattice

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Abstract. The collective charge state (holon) in a periodical lattice is defined. The exact commutation relations for the holon operators correspond to the modified para-Fermi statistics of rank M (M is the number of lattice sites at which the hole can be created), i.e. one state can be occupied by up to M holons. At small concentration, the holon gas can be treated as a bosonic system. In the general case, the holon gas is characterized by the immanent interaction and coupling among the different holon band states, even in the absence of dynamic interactions (the so-called kinematic interaction). In spite of the kinematic interaction, there is no statistical prohibition of Bose–Einstein condensation in the holon system. The possible extension of the approach developed to the coupled hole pairs in high-T_c superconductors is discussed.

1. Introduction: collective charge state

It is well established that in the high-T_c cuprates the charge carriers are positive. The study of the hole migration process has a great importance for elucidating the mechanism of high-T_c superconductivity; see the review by Dagotto [1]. The efficiency of charge transfer also determines properties of organic conductors and semiconductors, and irradiated polymers.

When a hole is created in some atom in a lattice or in a monomer in a polymer, it can migrate in a lattice or along a polymer chain. There is an equal probability for the location of the hole at each site of the same nature. As a result, the collective state is formed. This collective state can be considered as a charge wave or as a quasiparticle. In our paper the statistics of such quasiparticles at arbitrary concentrations is studied. For this purpose, we use the second-quantization formalism.

Usually holes are considered as fermion particles with spin $1/2$ and positive charge. In the second-quantization formalism, the hole operators are adjoint to the electron operators and obey the fermion commutation relations; see [2–4]. But in real systems holes are located on many-electron atoms or molecules and can have different values of spin S. For example, holes in the CuO_2 planes in high-T_c cuprate oxides have $S = 0$; this is the so-called Zhang–Rice singlet [5, 6]. In this study we consider holes as positive charged atoms (molecules) with $S = 0$, or as spinless quasiparticles in the second-quantization formalism. In the absence of dynamical interaction between holes, the model Hamiltonian for an arbitrary lattice with one type of hole can be written as follows:

$$H = \varepsilon_0 \sum_n b_n^+ b_n + \sum_{n,n'} M_{nn'} b_n^+ b_{n'} \tag{1}$$

where b_n^+, b_n are the hole creation and annihilation operators, respectively; ε_0 is the energy of the hole creation in a lattice and $M_{nn'}$ is the so-called hopping integral, characterizing the efficiency of charge transfer (hopping) from site n to site n'. According to the theory of resonance interaction (see section 1.2.3 in [7]), the hopping integral can be expressed in terms of the resonance integral in the form

$$M_{nn'} = \langle \Psi_0(A_{n'}^+)\Psi_0(A_n)|V_{int}|\Psi_0(A_{n'})\Psi_0(A_n^+)\rangle \tag{2}$$

where $\Psi_0(A_n)$ and $\Psi_0(A_n^+)$ are the ground-state many-electron wave functions for neutral and ionized atoms (monomers) located at site n. It can be shown (we will do it elsewhere) that in the case of a one-electron wave function expression (2) has the same physical sense as the hopping integral in the Hubbard Hamiltonian; cf. reference [8].

If the wave functions in equation (2) are not overlapping, the operators $b_{n'}^+$, b_n acting on different sites must commute. Thus, they obey the Bose commutation relations:

$$\left[b_n, b_{n'}^+\right]_- = [b_n, b_{n'}]_- = \left[b_n^+, b_{n'}^+\right]_- = 0 \qquad \text{for } n \neq n'. \tag{3}$$

In the Hamiltonian given by equation (1) it is assumed that at a site only one hole can be created, and that of only one type. Although the definition of the hole is quite general and it can correspond to an arbitrary n-fold ionized state, we do not consider states with two holes at one site. From this it follows that

$$(b_n^+)^2|0\rangle = 0 \tag{4}$$

where $|0\rangle$ is the vacuum state. As the vacuum state, we consider a state in which all sites of a lattice are neutral. The operators acting on one site satisfy the Pauli principle and the Fermi commutation relations:

$$\begin{aligned} \left[b_n, b_n^+\right]_+ &= 1 \\ [b_n, b_n]_+ &= \left[b_n^+, b_n^+\right]_+ = 0. \end{aligned} \tag{5}$$

The operators with the commutation relations (3)–(5) are called the Pauli operators [9] and describe paulion particles. Thus, the operators introduced for holes are paulions like the second-quantization operators for spin [10] and electronic excitations in crystals [9] or the Cooper pair operators [11]. But in the latter case there is an essential difference: unlike the Cooper pair operators which are delocalized, the hole operators are localized at lattice sites. In this aspect, they are similar to spin operators and electronic excitations in a crystal.

The Hamiltonian in equation (1) can be diagonalized by some unitary transformation:

$$B_q = \frac{1}{\sqrt{M}} \sum_n u_{qn} b_n \qquad B_q^+ = \frac{1}{\sqrt{M}} \sum_n u_{qn}^* b_n^+ \tag{6}$$

where M is the number of lattice sites at which the hole can be created. With the new operators, the Hamiltonian is transformed from the site representation to the quasimomentum representation in which it has the diagonalized form

$$H = \sum_q \varepsilon_q B_q^+ B_q. \tag{7}$$

The action of the operator B_q^+ on the vacuum state $|0\rangle$ creates a collective charge state with a hole distributed among all equivalent sites. This state can be considered as a quasiparticle similar to magnon or exciton quasiparticles [9, 10], so it is natural to call it a *holon*. For a lattice with one atom (molecule) per cell, the holon energy is given by the following expression:

$$\varepsilon_q = \varepsilon_0 + \sum_{n'\,(\neq n)} M_{nn'} \exp[i\boldsymbol{q} \cdot (\boldsymbol{r}_n - \boldsymbol{r}_{n'})]. \tag{8}$$

It is important to stress that the collective charge state which we named a holon is different from the Anderson holon [12–15]. The latter is a much more specific object. It is described as the charged spinless boson which exists always with the fermionic spin soliton (spinon). In the Anderson model, the holon and spinon correspond to the charge and spin degrees of freedom of electrons which can be separated (see the interesting discussion in reference [16]). As is shown in our paper, the introduced holons are bosons only in the low-concentration limit; at large concentration they obey the modified para-Fermi statistics.

In 1953 Green [17] and, independently, Volkov [18] showed that the bosonic and fermionic commutation relations do not exhaust all physically possible commutation relations for field operators in the second-quantization formalism. The field operators satisfying the requirements of causality, relativistic invariance and positivity of energy can obey more general commutation relations than the bosonic and fermionic ones, namely

$$
\begin{aligned}
\left[\left[a_k^+, a_{k'}\right]_\pm, a_{k''}\right]_- &= -2\delta_{kk''}a_{k'} \\
\left[\left[a_k^+, a_{k'}\right]_\pm, a_{k''}^+\right]_- &= 2\delta_{k'k''}a_k^+ \\
\left[\left[a_k^+, a_{k'}^+\right]_\pm, a_{k''}^+\right]_- &= \left[\left[a_k, a_{k'}\right]_\pm, a_{k''}\right]_- = 0.
\end{aligned}
\tag{9}
$$

The relations (9) with the upper sign at the inner brackets are called the parabosonic commutation relations and those with the lower sign are called the parafermionic commutation relations. These names are connected with the fact that the ordinary bosonic and fermionic operators also obey the relations (9). But these relations are fulfilled by a more general type of operator given by the so-called Green *ansatz* [17]:

$$
a_k = \sum_{\rho=1}^{p} d_{\rho k}
\tag{10}
$$

with operators $d_{\rho k}$ obeying the following commutation relations:

$$
\begin{aligned}
\left[d_{\rho k}, d_{\rho k'}^+\right]_\mp &= \delta_{kk'} \\
\left[d_{\rho k}, d_{\rho k'}\right]_\mp &= \left[d_{\rho k}^+, d_{\rho k'}^+\right]_\mp = 0 \\
\left[d_{\rho k}, d_{\rho' k'}^+\right]_\pm &= \left[d_{\rho k}, d_{\rho' k'}\right]_\pm = \left[d_{\rho k}^+, d_{\rho' k'}^+\right]_\pm = 0 \qquad \text{for } \rho \neq \rho'.
\end{aligned}
\tag{11}
$$

The upper sign is for the parabosons; the lower sign is for the parafermions. We see that the operators $d_{\rho k}$ and $d_{\rho k}^+$ in the parafermionic case are the fermionic operators at the same ρ, and they are bosonic operators at different ρ (the picture is similar in the parabosonic case).

The value of p in equation (10) is called the rank of the parastatistics and corresponds to the maximum number of particles which can occupy one state. If $p = 1$, the parastatistics turns into the Fermi–Dirac statistics; for $p \to \infty$ it turns into the Bose–Einstein statistics. The statistical properties and thermodynamic functions of an ideal gas of paraparticles are presented in Isihara's book [19].

Although, all elementary particles known at present are bosons or fermions, the parastatistics can be realized for quasiparticles. As was shown by one of the authors [20], the quasiparticles in a periodical lattice (the Frenkel excitons and magnons) obey a modified para-Fermi statistics of rank M, where M is the number of equivalent lattice sites within the delocalization region of collective excitations. Later on, the results [20] for the Frenkel excitons and magnons were extended to polaritons [21,22], defectons in quantum crystals [23] and to the Wannier–Mott excitons [24]. Below, we study the statistics and some properties of a system of non-interacting holons following the approach developed in reference [20].

2. Statistics and some properties of holon gas

The holon operators B_q, B_q^+ are connected with the hole operators b_n, b_n^+ by a unitary transformation (6), i.e.,

$$\frac{1}{M} \sum_q u_{qn'}^* u_{qn} = \delta_{nn'}. \tag{12}$$

Since the hole operators obey neither the boson nor the fermion commutation relations, the unitary transformation in the general case is not canonical; this means that it does not preserve the commutation properties of the operators transformed. In fact, using the commutation relations for the Pauli operators, equations (3)–(5), the following commutators hold:

$$\left[B_q, B_{q'}^+\right]_- = M^{-1} \sum_n u_{qn} u_{q'n}^* (1 - 2b_n^+ b_n) \tag{13}$$

$$\left[B_q, B_{q'}^+\right]_+ = M^{-1} \sum_n u_{qn} u_{q'n}^* + 2M^{-1} \sum_{n,n'}' u_{qn} u_{q'n'}^* b_{n'}^+ b_n \tag{14}$$

where the prime on the summation sign means that $n \neq n'$.

For a simple periodical lattice with one atom (molecule) per cell, the unitary transformation that diagonalizes the Hamiltonian is completely determined by the translational symmetry of the lattice; in such a case u_{qn} in equation (6) is given by

$$u_{qn} = \exp(-i q \cdot r_n). \tag{15}$$

Therefore, we obtain

$$\left[B_q, B_{q'}^+\right]_- = \delta_{qq'} - \frac{2}{M} \sum_n \exp\{i(q' - q) \cdot r_n\} b_n^+ b_n \tag{16}$$

$$\left[B_q^+, B_{q'}^+\right]_- = \left[B_q, B_{q'}\right]_- = 0 \tag{17}$$

and

$$\left[B_q, B_{q'}^+\right]_+ = \delta_{qq'} + \frac{2}{M} \sum_{n,n'}' \exp\{i(q' \cdot r_{n'} - q \cdot r_n)\} b_{n'}^+ b_n. \tag{18}$$

The sums on the right-hand side of commutators (16) and (18) disappear if we calculate the trilinear commutators as in the parastatistics. For the para-Fermi case we obtain

$$\left[\left[B_q^+, B_{q'}\right]_-, B_{q''}\right]_- = -2M^{-1} B_{\bar{q}} \qquad \bar{q} = q' + q'' - q \tag{19}$$

$$\left[\left[B_q^+, B_{q'}\right]_-, B_{q''}^+\right]_- = 2M^{-1} B_{\bar{q}}^+ \qquad \bar{q} = q - q' + q''. \tag{20}$$

The relations (19) for $q = q''$ and (20) for $q' = q''$ are, within a normalization factor M, identical with the commutation relations for parafermions (see equations (9)). But for arbitrary q, q' and q'' there is one essential difference from the parafermion relations. In the latter there is a Kronecker symbol whereas in equations (19) and (20) the Kronecker symbol is absent; the state vector $\bar{q}$ on the right-hand side of (19) and (20) is determined by the quasimomentum conservation law. This leads to important physical consequences which we will discuss below.

To determine the action of the operator B_q on the vacuum state $|0\rangle$, the ground state at the Fermi level, it suffices to find the action of the Pauli operators on this state. The Pauli operators satisfy

$$b_n |0\rangle = 0 \qquad b_{n'} b_n^+ |0\rangle = \delta_{nn'} |0\rangle \tag{21}$$

and for the holon operators we obtain the same result:

$$B_q |0\rangle = 0 \qquad B_{q'} B_q^+ |0\rangle = \delta_{qq'} |0\rangle. \tag{22}$$

Using the definition of the operators B_q^+, equation (6), and the commutation relation for b_n^+, we can see that for the two-particle case

$$(B_q^+)^2|0\rangle = 2M^{-1} \sum_{n<n'} u_{qn'}^* u_{qn}^* b_{n'}^+ b_n^+ |0\rangle. \tag{23}$$

For the general case, $N < M$,

$$(B_q^+)^N|0\rangle = \left(\frac{N!}{M^{N/2}}\right) \sum_{n_1<n_2<\cdots<n_N} u_{qn_N}^* \cdots u_{qn_1}^* b_{n_N}^+ \cdots b_{n_1}^+ |0\rangle \tag{24}$$

and for the maximum value, $N = M$,

$$(B_q^+)^M|0\rangle = \left(\frac{M!}{M^{M/2}}\right) u_{qn_M}^* \cdots u_{qn_1}^* b_{n_M}^+ \cdots b_{n_1}^+ |0\rangle. \tag{25}$$

In the case $N = M + 1$ we will always have two particles at one site; therefore, using equation (4), we obtain

$$(B_q^+)^{M+1}|0\rangle = 0. \tag{26}$$

Thus, one state can be occupied by up to M quasiparticles. This means that holons satisfy some modified para-Fermi statistics of rank M. It is important to stress that this conclusion does not depend on the special choice of the unitary transformation (15) and is valid also for quasiparticles in a complicated lattice with several atoms (molecules) per cell. In the case of lattices with one atom (molecule) per cell the trilinear commutation relations transform to the simple form given by equations (19) and (20).

A function describing a state of N non-interacting particles, each with energy ε_q, is given by the usual expression

$$|N_q\rangle = C_N(B_q^+)^N|0\rangle. \tag{27}$$

To find the expression for the normalization factor it is convenient to use the following operator equation, which is obtained from equation (20):

$$B_q B_{q'}^+ B_{q''}^+ = B_{q'}^+ B_q B_{q''}^+ + B_{q''}^+ B_q B_{q'}^+ - B_{q''}^+ B_{q'}^+ B_q - \frac{2}{M} B_{\bar{q}}^+ \qquad \bar{q} = q' + q'' - q \tag{28}$$

or following from it

$$B_{q'}(B_q^+)^2 = 2B_q^+ B_{q'} B_q^+ - (B_q^+)^2 B_{q'} - \frac{2}{M} B_{\bar{q}}^+ \qquad \bar{q} = 2q - q'. \tag{29}$$

Using equation (29) and the induction method, we find the expression for the normalization factor, which differs from that for a Bose system:

$$C_N = \left[N!\left(1 - \frac{1}{M}\right)\left(1 - \frac{2}{M}\right)\cdots\left(1 - \frac{N-1}{M}\right) \right]^{-1/2}. \tag{30}$$

The results of applying the operators B_q^+ and B_q to the state vector $|N_q\rangle = C_{N_q}(B_q^+)^{N_q}|0\rangle$ are

$$B_q^+|N_q\rangle = \sqrt{(N_q + 1)\left(1 - \frac{N_q}{M}\right)}\,|N_q + 1\rangle \tag{31}$$

$$B_q|N_q\rangle = \sqrt{N_q\left(1 - \frac{N_q - 1}{M}\right)}\,|N_q - 1\rangle. \tag{32}$$

Equation (31) shows that the effect of applying B_q^+ on a state with a maximum occupation number $N_q = M$ is equal to zero. As $M \to \infty$, relations (31) and (32) turn into the well known relations for bosons.

From equations (31) and (32), it follows that

$$B_q^+ B_q |N_q\rangle = N_q \left(1 - \frac{N_q - 1}{M}\right)|N_q\rangle. \tag{33}$$

This means that the operator $B_q^+ B_q$ is not a particle number operator as in the case of boson, fermion or paulion operators. For the commutator we obtain

$$\left[B_q, B_q^+\right]_- |N_q\rangle = \left(1 - \frac{2N_q}{M}\right)|N_q\rangle. \tag{34}$$

Hence,

$$\frac{1}{2}M(1 - \left[B_q, B_q^+\right]_-)|N_q\rangle = N_q|N_q\rangle. \tag{35}$$

Thus, the operator of the particle number in state q is

$$\widehat{N}_q = \frac{1}{2}M(1 - \left[B_q, B_q^+\right]_-). \tag{36}$$

The trilinear commutation relation (20) can be expressed via the $\widehat{N}_q$ as

$$\left[\widehat{N}_q, B_{q'}^+\right]_- = B_{q'}^+. \tag{37}$$

The parafermionic commutation relation, because of the Kronecker symbol, gives for the particle number operator, instead of equation (37), the relation

$$\left[\widehat{N}_q, B_q^+\right]_- = B_q^+. \tag{38}$$

Equation (38) is the commutation relation which must be fulfilled by the operator of particle number: the action of the commutator (38) on the state $|N_q\rangle$ is equivalent to the increase of the eigenvalue of the operator $\widehat{N}_q$, while the commutation relation (37) leads to an unusual result: the eigenvalue of the operator $\widehat{N}_q$ depends upon the occupation number of state q' for all states q'. From this, it follows that the operator $\widehat{N}_q$ cannot belong only to the state q and has to be considered as the operator of the total number of quasiparticles. It is easy to show that this is indeed the case.

Substituting equation (16) into the expression (36) for $\widehat{N}_q$, we obtain

$$\widehat{N}_q = \frac{1}{2}M(1 - \left[B_q, B_q^+\right]_-) = \sum_n b_n^+ b_n = \widehat{N}. \tag{39}$$

Since the number of quasiparticles is equal to the number of holes at the sites, the operator (36) is the operator of the total number of quasiparticles, and it does not depend on q. From equation (39) it follows that

$$\left[B_q, B_q^+\right]_- = 1 - \frac{2\widehat{N}}{M}. \tag{40}$$

This means that, for $\langle \widehat{N} \rangle \ll M$, the quasiparticles satisfy the Bose statistics with good accuracy.

The Hamiltonian of an ideal gas of holons must be linear in the particle number operator:

$$H = \sum_q \varepsilon_q \widehat{N}_q. \tag{41}$$

As was shown above, we cannot introduce the holon number operator for a particular state because of the immanent coupling of different holon states even in the absence of dynamical interaction. Thus, it is impossible to describe an ideal gas of holons beyond the Bose approximation which is valid only for small concentration of holons. In general, the interaction between quasiparticles is always present. This kind of interaction depending on the deviation of

quasiparticle statistics from the Bose (Fermi) statistics is called, after Dyson [25], the *kinematic interaction*.

In order to estimate the magnitude of the kinematic interaction, it suffices to calculate the expectation value of the Hamiltonian (7) with the state vector (27):

$$E_{N_q} = \langle N_q | H | N_q \rangle = C_N^2 \langle 0 | B_q^N \sum_{q'} \varepsilon_{q'} B_{q'}^+ B_{q'} (B_q^+)^N | 0 \rangle. \tag{42}$$

To calculate this mean value it is convenient to use the operator equation (28) and the properties given by equation (22). As a result, for two quasiparticles with energy ε_q, we obtain

$$E_{2_q} = 2\varepsilon_q \left(1 - \frac{1}{M} \right) + \frac{2}{M-1} \frac{1}{M} \sum_{q' (\neq q)} \varepsilon_{q'} \tag{43}$$

and for the general case of N quasiparticles

$$\begin{aligned} E_{N_q} &= N\varepsilon_q \left(1 - \frac{N-1}{M} \right) + \frac{N}{M-1} \frac{N-1}{M} \sum_{q' (\neq q)} \varepsilon_{q'} \\ &= N \left[\varepsilon_q + \frac{N-1}{M} \left(\frac{1}{M-1} \sum_{q' (\neq q)} \varepsilon_{q'} - \varepsilon_q \right) \right]. \end{aligned} \tag{44}$$

The second terms in equation (44) contain the concentration of quasiparticles.

Equation (44) can be written as

$$E_{N_q} = N \left[\varepsilon_q + \frac{N-1}{M} (\bar{\varepsilon} - \varepsilon_q) \right] \tag{45}$$

where

$$\bar{\varepsilon} = \frac{1}{M-1} \sum_{q' (\neq q)} \varepsilon_{q'} \tag{46}$$

denotes the mean energy of the holon band. In equation (45) the second term describes the kinematic interaction, so the holon gas acquires a more ideal behaviour when the energy ε_q comes nearer to the mean energy $\bar{\varepsilon}$ of the band.

The number of holons is equal to the number of created holes. The latter cannot exceed the number of lattice sites M at which the hole can be created. So, all created holons can occupy one state, for example the ground state. This means that, in spite of the non-bosonic behaviour of the holon gas, there is no statistical prohibition of the Bose–Einstein condensation phenomenon in holon systems. On the other hand, the holon gas is always non-ideal (because of the kinematic interaction). The study of the Bose–Einstein condensation in non-ideal systems requires a special treatment of the stability of the Bose condensate [26]. For rigorous study of this problem we must also include a dynamic interaction and consider an interplay between kinematic and dynamic interaction in the holon system, as was done for the molecular exciton system in reference [27].

The method developed here can be extended to study the system of coupled holes in high-T_c superconducting ceramics. In this case it is convenient to use the Hubbard model with the coupled interaction term—this means the hole–hole attractive interaction, which could be simple and general because it does not depend explicitly on the nature of interactions between holes; they are included as parameters in the Hamiltonian. The coupling could also have s-wave nature as in the BCS case or d-wave nature [28]. It is well known that the positive pairs in high-T_c ceramics have localized nature; the correlation length in CuO_2 planes is of the order of 10–12 Å. To reveal the statistics of the hole pairs a special study is needed, though we can expect the system of such coupled localized hole pairs to obey some modified para-Fermi

statistics like the holon system studied, but with the rank $M' = M/m$, where m is the number of sites in the localized region of two coupled holes. So, the number of possible pairs cannot be larger than the rank of the parastatistics describing the coupled hole pairs. This means that there are no statistical hindrances to the Bose–Einstein condensation of hole pairs. Note that for the low-hole-density limit (the one-pair problem), we obtain the same result for the binding energy as in the Cooper pair case. Details of this problem and a study of the many-pair problem that is under way will be published elsewhere.

3. Conclusions

We have studied the statistical properties of the collective charge states (holons) in a periodical lattice using the exact trilinear commutation relations for the holon operators. As was shown, the holons obey the modified para-Fermi statistics of rank M, where M is the number of lattice sites at which the hole can be created, i.e. one state can be occupied by up to M holons. The number of holons is equal to the number of holes and cannot be larger than the number M of sites. Thus, there is no statistical prohibition of the Bose–Einstein condensation phenomenon in holon systems.

The second important conclusion of this study is that, in general, the system of dynamically non-interacting holons cannot be considered as an ideal gas. In the holon system the immanent interaction is always present and so is the coupling of all states of the holon band (the so-called kinematic interaction). The magnitude of the kinematic interaction is proportional to the holon concentration, N/M, and has the same order as the corrections for the non-Bose behaviour in the commutation relations for the holon operators. This means that the holon gas can be considered as ideal only in the Bose approximation which is valid at small concentration.

The approach developed in this study can be extended to the system of coupled holes in high-T_c superconducting ceramics, since it is well known that the positive pairs in high-T_c ceramics have a localized nature with a correlation length in CuO_2 planes of the order of 10–12 Å. Thus, there is no statistical prohibitions of the phenomenon of Bose–Einstein condensation of coupled hole pairs, although to establish the Bose–Einstein mechanism it is necessary to perform a study of the stability of the Bose condensate.

Acknowledgments

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