TWO DOPED CARRIER BOUND STATES DUE TO SPIN CONSTRAINT IN ANTIFERROMAGNETS

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UDC 539

It is shown that two nearest neighbor doped carriers in antiferromagnets with the Néel ground state can freely propagate along a crystal as opposed to single carriers.

The problem of metallization of initially nonconducting crystals (mainly semiconductors) upon their doping may be considered as solved (see, e.g., [1–3]). However, the discovery of high-temperature superconductors (HTSCs) raised a problem on the nature of conductivity in strongly correlated fermion systems and renewed an interest to properties of doped materials. In his first paper on HTSCs [4] Anderson called attention to the fact that the conducting elements in these materials are the cuprate layers CuO_2 with the copper subsystem being described by the Hubbard model on a square lattice with the Mott—Hubbard parameter $U/t \gg 1$, where t is the hopping amplitude and U is the Hubbard on-site repulsion (see also [5, 6].

At half-filling, the band structure of CuO_2 layers for large values of the Mott-Hubbard parameter consists of two Hubbard subbands one of which is empty and another is fully occupied. In terms of the tight binding approach, it corresponds to the antiferromagnetic (AFM) Heisenberg model of spins $S = 1/2$ on a square lattice with the exchange integral $J \sim t^2/U$ between nearest neighbors. In the limit $U/t \gg 1$, the appearance of doubly occupied sites (or in other words, the appearance of electrons in the upper subband) becomes energetically unfavorable, and the model has an insulating Néel-ordered ground state [7, 8]. Doping such systems means either pulling electrons out of the filled Hubbard subband and providing the hole conductivity of the system or adding excess electrons and, in this way, creating a certain amount of doubly occupied sites — "twos" in the lattice (the electron conductivity along the upper Hubbard subband). Both doping processes are physically equivalent and, in what follows, we restrict our consideration for the sake of definiteness to the latter case. In principle, excess electrons (holes) may propagate along the lattice. However, there is a serious hindrance to such a motion caused by a spin constraint. The physical reason for this constraint is the following. In antiferromagnets with the classical Néel ground state (*i.e.* without regard for small quantum spin fluctuation effects which are of the order of $|v|^2 \ll 1$, where v is the coefficient of the Bogolyubov–Tyablikov uv-transform [9]), each electron transfer from one site to the nearest neighbor site is accompanied by the destruction of the Néel magnetic structure and by the loss in energy $\sim J$. The energy loss increases with the number of jumps [10, 11]. In fact, this means that electrons (holes) are localized. On the other hand, in the frame of the Hubbard model $t \gg J$ and in accordance with the Nagaoka theorem [12], the motion of electrons must lead to the creation of regions with ferromagnetic order in crystals [13] which have not been observed in HTSCs, to the best of our knowledge.

The applicability of the one-band Hubbard model to CuO_2-layers of HTSCs was questioned in [10, 14–16]. In the cited papers (see also a very comprehensive discussion of this problem in the review article [7],) a three-band model was developed, which takes into

account that excess carriers can occupy both copper and oxygen states of the cuprate layers. However, this model is rather complicated and the most commonly used model is the so-called tJ-model [17] with a Hamiltonian in the form

$$H_{tJ} = \sum_{\vec{n}\vec{m}} \left(- \sum_{\sigma} t_{\vec{n}\vec{m}} \tilde{a}^{\dagger}_{\vec{n}\,\sigma} \tilde{a}_{\vec{m}\,\sigma} + J_{\vec{n}\vec{m}} \vec{S}_{\vec{n}} \cdot \vec{S}_{\vec{m}} \right), \qquad (1)$$

where $a^{\dagger}_{\vec{n}\,\sigma}$ ($a_{\vec{n}\,\sigma}$) creates (annihilates) an electron with the spin projection σ on site $\vec{n}$, the operator $\tilde{a}^{\dagger}_{\vec{n}\sigma}$ is equivalent to the Hubbard operator $X^{2\sigma}_{\vec{n}}$ which describes the creation of an electron with the spin $-\sigma$ at $\vec{n}$ which has already been occupied by an electron with the spin σ [18] $\vec{S}_{\vec{n}} = \frac{1}{2} \sum_{\sigma,\sigma'} a^{\dagger}_{\vec{n}\,\sigma} \hat{\vec{\sigma}}\, a_{\vec{n}\,\sigma'}$ is the spin operator, where the vector $\hat{\vec{\sigma}} = (\hat{\sigma}_x, \hat{\sigma}_y, \hat{\sigma}_z)$ is constructed from the Pauli spin matrices. It is worth stressing that the hopping amplitude t and the exchange integral J in Hamiltonian (1) are considered as independent. The model given by Hamiltonian (1) is one of the fundamental models in the physics of magnetism [18]. In the framework of this model, one can study the excess carrier motion in AFM crystals[1].

Let us assume that there is an excess electron in a two-sublattice AFM crystal. The appearance of an electron in such a crystal means that four exchange bonds are broken. Therefore, the crystal with an extra electron has an excess energy $4JS^2$ (S is the mean spin on-site value for a given temperature). The electron transfer to the nearest neighbor is accompanied by the energy increase $3JS^2$. Each new electron jump adds JS^2 and therefore a "string" of broken spins is coming into play [11] with the energy proportional to its length. The appearance of such a string means that the motion of a carrier is energetically unfavorable and this provides its actual localization. The situation reverses when the AFM crystal contains at least two carriers. In this case, as will be shown below, the bound state of two carriers is essentially delocalized.

Let us consider the problem of eigenstates and eigenvalues for two doped electrons in a two-sublattice Néel AFM. For the sake of simplicity we restrict ourselves to the case of an Ising antiferomagnet and choose the Hamiltonian of the system as

$$H = H_{\rm Ising} + \epsilon H_{\rm hop}, \qquad (2)$$

where the term

$$H_{\rm hop} = - \sum_{\vec{n}\vec{m}} \sum_{\sigma} t_{\vec{n}1\,\vec{m}2} \left(\tilde{a}^{\dagger}_{\vec{n}1\,\sigma} \tilde{a}_{\vec{m}2\,\sigma} + {\rm h.c.} \right), \qquad (3)$$

describes the dispersion of carriers and the term

$$H_{\rm Ising} = \sum_{\vec{n}\vec{m}} J_{\vec{n}1\,\vec{m}2}\, S^z_{\vec{n}1} S^z_{\vec{m}2} \qquad (4)$$

gives the exchange interaction between atoms in the Ising approximation. We are interested in the case of narrow-band crystals, $t < J$. To emphasize this, a formal small parameter ϵ was introduced in Eq. (2). The notation $\vec{n}\alpha$ means that the corresponding site belongs to the sublattice α ($\alpha = 1, 2$) in the elementary cell $\vec{n}$.

In the ground state of an undoped crystal which we denote as $|{\rm Neel}\rangle$, the first sublattice $\alpha = 1$ is created from spins "up" ($\sigma = \uparrow$). In the second sublattice $\alpha = 2$, all spins look "down" ($\sigma = \downarrow$). Then the wave function

$$\Psi_{\vec{n}\vec{m}} = a^{\dagger}_{\vec{n}1\downarrow} a^{\dagger}_{\vec{m}2\uparrow} |{\rm Neel}\rangle \qquad (5)$$

corresponds to two excess electrons in the crystal. In the no-hopping limit (zero-approximation with respect to ϵ), the level $E_0 = -7JS^2$ is degenerated because all functions (5), $\vec{n}1$ and $\vec{m}2$ being the nearest neighbors, have this energy. Denoting the corresponding linear space by L and introducing the projection operator P to this space, one can write

$$H_{\rm Ising} P\Psi = E_0\, P\Psi. \qquad (6)$$

To study the role of the hopping term (3), we use the perturbation theory for degenerated levels which was developed by Bogolyubov [20]. In the second order approximation, the eigenfunctions and energy eigenvalues of the AFM with two doped electrons are determined by the equation

$$(E - H_{\rm eff})\, P\Psi = 0, \qquad (7)$$

where the second-order effective Hamiltonian has the form

$$H_{\rm eff} = H_{\rm Ising} + \epsilon P\, H_{\rm hop}\, P + $$

$$+ \epsilon^2 P\, (H_{\rm hop} - P\, H_{\rm hop}\, P\,)\, \frac{1}{E_0 - H_{\rm Ising}} \times$$

$$\times\, (H_{\rm hop} - P\, H_{\rm hop}\, P\,)\, P. \qquad (8)$$

[1] Note that such a kind of models is well known and widely used in the physics of magnetic excitons, where the band widths of excitons and magnons given by t and J are independent parameters (see, e.g., [19]).

It is easy to see from the definition of the linear space L that $P\,H_{\rm hop}\,P=0$ and

$$\epsilon^2\,P\,H_{\rm hop}\,\frac{1}{E_0-H_{\rm Ising}}\,H_{\rm hop}\,P=\frac{1}{\Delta E}\times$$

$$\times P\sum_{\vec{n},\vec{m},\vec{l}}\Big\{t_{\vec{n}1\,\vec{m}2}\,t_{\vec{l}1\,\vec{m}2}\,(1-\delta_{\vec{n}\,\vec{l}})\,a^{\dagger}_{\vec{m}2\downarrow}\,a_{\vec{n}1\downarrow}\,a^{\dagger}_{\vec{l}1\downarrow}\,a_{\vec{m}2\downarrow}+$$

$$+t_{\vec{n}1\,\vec{m}2}\,t_{\vec{n}1\,\vec{l}2}\,(1-\delta_{\vec{m}\,\vec{l}})\,a^{\dagger}_{\vec{n}1\uparrow}\,a_{\vec{m}2\uparrow}\,a^{\dagger}_{\vec{l}2\uparrow}\,a_{\vec{n}1\uparrow}\Big\}\,P, \qquad (9)$$

where $\Delta E=3JS^2$ is the energy difference between the state with two nearest neighbor carriers and the state with two well separated carriers. The Kronecker deltas in Eq. (9) account for the kinematic repulsive interaction between carriers.

Representing the eigenfunction of the AFM with two doped electrons in the form

$$P\Psi=\sum_{\vec{n}\,\vec{m}}C_{\vec{n}\,\vec{m}}\,\Psi_{\vec{n}\,\vec{m}}, \qquad (10)$$

where the double sum is limited to pairs of nearest neighboring sites, we obtain the equations for coefficients $C_{\vec{n}\vec{m}}$ from Eqs. (4), (7)—(9) in the form

$$(E-E_0)\,C_{\vec{n}\vec{m}}=\frac{1}{\Delta}\sum_{\vec{l}}\Big\{t_{\vec{n}1\,\vec{m}2}\,t_{\vec{l}1\,\vec{m}2}\,(1-\delta_{\vec{n}\,\vec{l}})\;C_{\vec{l}\vec{m}}+$$

$$+t_{\vec{n}1\,\vec{m}2}\,t_{\vec{n}1\,\vec{l}2}\,(1-\delta_{\vec{m}\,\vec{l}})\;C_{\vec{n}\vec{l}}\Big\}. \qquad (11)$$

It is convenient to introduce the center-of-mass coordinate $\vec{R}=(\vec{n}+\vec{m})/2$ and the coordinate of the relative motion $\vec{r}=\vec{n}-\vec{m}$. Then, for the Fourier transform

$$C_{\vec{K}}(\vec{r})=\frac{1}{\sqrt{N}}\sum_{\vec{R}}e^{i\,\vec{K}\cdot\vec{R}}\,C_{\vec{R}+\vec{r}/2,\,\vec{R}-\vec{r}/2}, \qquad (12)$$

we obtain from Eq. (11) that

$$(E-E_0)\;C_{\vec{K}}(\vec{r})=\frac{2t}{\Delta}\sum_{\vec{\rho}}\;(1-\delta_{\vec{r}\vec{\rho}})\;t_{\vec{\rho}1\vec{0}2}\times$$

$$\times\cos\left(\frac{1}{2}\,\vec{K}\cdot(\vec{r}-\vec{\rho})\right)\;C_{\vec{K}}(\vec{\rho}). \qquad (13)$$

In accordance with the crystal group symmetry (D_{4h}), there are two types of eigenstates: totally symmetric A_g states and d-like B_g states. Their energies are given by the expressions

$$E_{\pm}(\vec{K})=-JS^2+$$

$$+2\,\frac{t^2}{\Delta}\left[1\pm2\cos\left(\frac{K_x a}{2}\right)\,\cos\left(\frac{K_y a}{2}\right)\right], \qquad (14)$$

where $+(-)$ corresponds to the $A_g\,(B_g)$-state. In Eq. (14), the energy is measured from the energy of two well separated carriers and a is the distance between the nearest neighbor sites. Thus, we can conclude that

- it is energetically favorable for electrons (holes) to unite into pairs.
- In contrast to single carriers, the bound states of two carriers from different AFM sublattices are delocalized ones.
- The binding energy of two carriers in the d-like B_g-state is higher than that in the totally symmetric A_g-state.
- The energy gain for the carrier pairing stems from the nature of the antiferromagnetic state of a crystal, and their existence does not require any dynamical attraction between carriers.

It is worth noting that the last two items are in full agreement with results of numerous numerical simulations (see, e.g., [7]) which showed that it is in the case $t<J$ the carriers in lightly doped AFMs tend to pair and to create the bound states which are non-totally symmetric. The possibility to create *mobile* pairs was indicated in the review paper [7] where the results of [21] were used. However, in contrast to us, the bound states of two carriers in an empty (non-AFM) lattice were considered in [21].

In conclusion, we have investigated the pairing of carriers (electrons or holes) in the antiferromagnetic tJ-model. It was shown that, for $t<J$, the bound states of two dynamically non-interacting electrons (holes) may exist only due to a *spin constraint* caused by the nature of the antiferromagnetic Néel ground state. With a rise of carrier density and/or temperature when neighboring spins are not completely antiparallel, the tendency to pairing decreases. It is evident that such a pairing exists in a rather narrow range of the parameters t and J. On the one hand, it is absent for $t>J$ and, on the other hand, it ceases to exist as the ratio J/t increases because stripe phases become more energetically favorable in this case [7].

We are indebted to S.M. Ryabchenko and Yu. Pogorelov for discussions. one of us (Y.B.G.) gratefully acknowledges the partial support from Deutsche Zentrum für Luft- und Raumfart e.V., Internatioles Büro des Bundesministeriums für Forschung und Technologie, Bonn, in the frame of a bilateral scientific cooperation between Ukraine and Germany, project No. UKR-02/011.

Note added in proof: (March, 31, 2005) After this paper had been submitted, we learned about the article by A.F. Andreev (ZhETF, Pis'ma, **79**, 100 (2004)) where the problem on possible bound states of two electrons in antiferromagnetic square lattice was also investigated. Here the two-electron bound states are considered in the belief that there is an intra-sublattice electron transfer (or in other words, there is an electron hopping to the next-neighbor sites). Contrary to this we neglected the next-nearest-neighbor electron hopping for reasons of smallness of the corresponding hopping parameter. It is worth noting that the problem on bound state of two electrons from different magnetic sublattices with the allowance of intra-sublattice electron transfer is identical to the problem on biexciton motion in antiferromagnetic dielectrics (see [19]).

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Received 08.08.04

ЗВ'ЯЗАНІ СТАНИ ДВОХ ДОПОВАНИХ НОСІЇВ, ЗУМОВЛЕНІ СПІНОВИМ ЗВ'ЯЗКОМ У АНТИФЕРОМАГНЕТИКАХ

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Р е з ю м е

Показано, що в антиферомагнетиках з неєлівським основним станом два сусідні доповані носії можуть вільно поширюватись вздовж кристала на відміну від поодиноких носіїв.