Physica A 168 (1990) 705–713
North-Holland

PHASE COHERENCE OSCILLATION OF HOLES IN $La_{2-x}(Sr)_xCuO_4$, DYNAMICS OF SINGLE HOLES IN THE CuO PLANE AND THE TYPICAL PAIRING TIME

Raphael BLUMENFELD
Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge CB3 0HE, England

Received 6 July 1990

Following the recently suggested vortex/antivortex-mediated pairing mechanism in $La_{2-x}(Sr)_xCuO_4$, it is proposed here, as a possibility, that single holes, induced by low doping, reside in a symmetric double well potential (DWP), formed by the vortex/antivortex excitation. An oscillatory behaviour is then predicted, originating from the oscillation in the probability to find the hole in either of the potential wells. It is further shown that the existence of such a DWP excitation gives rise to a simple mechanism for the movement of a single hole within the antiferromagnetic background. The typical time for pairing is then calculated in terms of the barrier height of the DWP.

1. Introduction

It is generally accepted that the CuO plane in pure La_2CuO_4 high-temperature superconductors can be envisaged as composed of spins with antiferromagnetic couplings between any two neighbours, J_{AF} [1, 2]. By doping of Sr, to form $La_{2-x}(Sr)_xCuO_4$, holes are induced, which are roughly located on the planar oxygen atoms [3]. These holes are distinct from those on the copper atoms, which are responsible for the underlying antiferromagnetic background. The holes associated with the oxygen atoms are believed to be responsible for the supercurrent. It has been noted [4] that the net magnetic effect of the holes, induced by doping, is to produce an effective ferromagnetic interaction, J_F, between nearest copper atom neighbours. Recently two independent groups carried out analytic [5] and numerical [6] studies that resulted in the suggestion that the pairing between holes in the high-temperature superconductors is mediated by a vortex/antivortex attraction. Both groups assumed that the antiferromagnetic background consists of spins that can rotate in the plane (XY spin model) and showed that the frustrating holes act as nucleation

centers around which a vortex (or an antivortex) is very likely to be excited. Once a vortex is excited around a hole (H-vortex), it creates a region of locally high energy and favours excitation of a spontaneous (S-)antivortex to form a bound pair, SH, and to reduce the energy. The probability of such a spontaneous excitation to occur increases below the Kosterlitz–Thouless temperature. Hence, at sufficiently low temperatures, we may assume that a substantial fraction of all holes are in such a SH configuration. Such a bound pair of an excitation centered around a hole and its counterpart without a hole, is very similar to the spinon–holon excitation that occurs in the fractional charge model [7], thus making it possible that the following analysis applies to that model as well. Numerical simulation [6] showed that once two SH excitations are near enough, they attract and are most likely (for temperatures below the transition) to form a bound state with one hole residing within the vortex and the other within the antivortex – a HH state. This state was found to be more favourable energetically than two nearby pairs of SH excitations. Thus an effective attraction between the holes is generated, mediated by the vortex/antivortex bound state. This pairing mechanism is very appealing in its simplicity and some of its implications are the subject of the present paper. Again, owing to the similarity in the geometrical configuration, one can naively parallel this HH state to the holon–holon excitation in the fractional charge model. Adding to these similarities the resemblance between the spinon–spinon and spontaneous vortex/antivortex excitations that occur without any hole (the usual Kosterlitz–Thouless mechanism [8]), SS, it is very tempting to conjecture, as done in ref. [6], the existence of some formal mapping between the two models. Such a mapping, if it exists, is interesting mainly due to the fact that the fractional charge model is presently believed to have no classical analog.

The main purpose of this paper is to study: (i) the SH excitation more explicitly, and (ii) the dynamics that leads two such excitations to move around in the antiferromagnetic plane, until they combine to form a bound HH pair. In the first part of the paper, section 2, it is proposed that in the SH state the hole resides in an effective double well potential (DWP). Hence, by applying known textbook results for the behaviour of a particle in such a potential, conclusions can be drawn concerning the present situation. The primary conclusion reached here is that there appears an oscillatory behaviour that corresponds to the temporal periodicity in the probability of the hole to be observed in either of the wells. This result has experimental implications: (i) it makes the basic suggestion susceptible to examination and (ii) it links the frequency of oscillation to the height of the barrier between the wells, enabling the direct examination of this parameter. In the second part of the paper, section 3, it is proposed that as the temperature increases, but is still below the

transition, the DWP shape of the SH excitation allows for a novel mechanism for the dynamics of the single hole in the antiferromagnetic background. The effective movement of the hole is then mapped into an equivalent random walk with a waiting time distribution that relates to the parameters of the DWP. From that analysis we can deduce the typical "mean free" time for combination of two SH excitations into a HH. This typical time is then related to the height of the barrier between the wells in the DWP. Comparing with the analogous parameter extracted from the frequency of the oscillations described in the first part, the variation of the height with temperature can be studied. This comparison may also serve as a check on the suggestions presented here. In section 4 we discuss the results.

2. The ground states of a SH excitation and typical oscillatory behaviour

In the following we consider the regime of low doping and low temperature, such that there already exist, albeit sparsely distributed, SH excitations. As suggested in ref. [6], the hole may be viewed as rotating within a vortex that occupies one plaquette of size $a \times a$, where a is the lattice parameter. The CuO plane is assumed to consist of $(L/a) \times (L/a)$ plaquettes in all. In the absence of higher excitations of the SH that are close in energy to its ground state (in the range of temperatures discussed here this is most likely to be the case), the hole sees an effective DWP with one vortex, representing a well, centered around the hole and the other, a spontaneous antivortex representing the second well, is located in the immediate vicinity. In the dilute limit the interaction with further vortices is very weak and for the moment we disregard it altogether. We will return to this point in the concluding discussion to determine how low the concentration of holes should be for this condition to hold. The energy of the configuration when the hole resides in the vortex, is the same as when it resides in the antivortex, implying that this state is doubly degenerate. It follows that the DWP that the hole sees is symmetric, with a spatial separation of la between the wells, and where l is some number of order 1. For the moment we assume that the DWP is fixed in space, which implies assumptions on the typical times involved in the physical system and on the temperature; points that we defer to the concluding discussion. The barrier height of the DWP can be naively estimated by removing the hole to infinity and observing that the interaction between two Cu atoms changes by $|J_{AF}| + |J_F|$. The behaviour of a particle in a symmetric DWP is reasonably well understood [9], and we will only outline briefly some of the features relevant to the present analysis. The system can be schematized by assuming it has two *non-orthogonal* "left" and "right" configurations that correspond to the hole

residing in the vortex, $|V\rangle$, and in the antivortex, $|AV\rangle$. The fact that the higher energy levels are unattainable, enables to consider, to a good approximation, only a two-dimensional Hilbert space. This is also tantamount to restricting the range of temperatures from above, such that degrees of freedom like translation and rotation (see below for the mechanism to invoke such motions) of this complex in the CuO plane are either too slow, or nonexcitable at all. This condition is consistent with the current phase diagram of La_2CuO_4, since for low temperatures and low enough concentrations, these materials are known to exhibit a spin-glass type of behaviour. Namely the holes may be considered almost completely quenched. Due to the symmetry, both configurations then have the same energy,

$$\langle V|H|V\rangle = \langle AV|H|AV\rangle \equiv E_0 , \tag{1}$$

and we also have

$$\langle V|V\rangle = \langle AV|AV\rangle = 1 .$$

Since they are not orthogonal their overlap integral is finite,

$$\langle V|AV\rangle \equiv \varepsilon , \tag{2}$$

and it is assumed to be proportional to the matrix element in the Hamiltonian that couples the two states,

$$\langle V|H|AV\rangle \equiv (E_0 + \delta E)\varepsilon . \tag{3}$$

Thus the Hamiltonian can be written as a sum of two Pauli's spin matrices, σ_z and σ_x. The diagonalization of such a Hamiltonian presents no problem and the two eigenvalues are

$$E_i = E_0 \pm \frac{\varepsilon\,\delta E}{1 \pm \varepsilon} , \tag{4}$$

which correspond to a symmetric ($i = +$) and an antisymmetric ($i = -$) combination of $|V\rangle$ and $|AV\rangle$,

$$|\Psi_\pm\rangle = \frac{1}{\sqrt{2(1 \pm \varepsilon)}} (|V\rangle \pm |AV\rangle) . \tag{5}$$

Correspondingly, E_+ and E_- are the energy of the lower and upper states. Thus the real ground state is stable and is composed of a symmetric combina-

tion of the hole being in both wells. To analyze the time behaviour of this system, assume that at time $t = 0$ an experiment is carried out in which the hole is observed within, say, the vortex. We wish to find the probability $p_v(t)$ that at a later time $t \neq 0$, the hole will still be observed within the same vortex, $|V\rangle$. As time passes, the system evolves into a mixed state $|\Phi(t)\rangle$ according to

$$|\Phi(t)\rangle = \sqrt{(1+\varepsilon)/2}\, e^{-i2\pi E_+ t/h}|\Psi_+\rangle + \sqrt{(1-\varepsilon)/2}\, e^{-i2\pi E_- t/h}|\Psi_-\rangle . \tag{6}$$

The probability $p_v(t)$ is then the square of the overlap between $|\Phi(t)\rangle$ and the initial state $|V\rangle$,

$$p_v(t) = |\langle V|\Phi(t)\rangle|^2 = [\varepsilon^2 + (1-\varepsilon^2)\cos^2\theta] , \tag{7}$$

where $\theta \equiv 2\pi\, \delta E\, t/h$. This probability oscillates with time, which is a spectacular consequence of the phase coherence between the amplitudes of being in the vortex and in the antivortex. A similar phenomenon occurs, e.g., in the ground state of an ionic molecule H_2^+, in optically active NH_3 and in the benzene molecule [9]. Note that although the low-temperature regime has been assumed, the spins are considered to be free enough to form vortices.

3. The transport mechanism of a single hole in the dilute regime

In addition to the above implications, the picture presented here suggests a novel mechanism for the transport of the hole within the antiferromagnetic background. In the foregoing the DWP has been considered stationary due to the low temperature and low concentration. However, in order to combine with another SH excitation, our complex must travel in the CuO plane. As the concentration increases, such a complex starts to unpin, and it seems plausible that the vortex with the hole, H, is less mobile than the spontaneous one, S. This combination of paired "heavy" and "light" vortices causes the latter to rotate and assume random positions around the former. This situations differs from the one considered above in the sense that we now allow other degrees of freedom. Although this may seem to disturb the assumptions that were introduced above, one should realize that the existence of a vortex / antivortex pair with a shared hole, makes it very plausible that the hole continues to see an effective DWP. In the following we simplify the picture by naively approximating the "massive" vortex, H, to be much heavier, so that it acts as a fixed pivot around which the lighter S changes position (in a more precise analysis, the "center of mass" and the "reduced mass" of the two excitations should be taken into account). We now define the probability $f(\boldsymbol{r})$ of the S to assume

position $\boldsymbol{r}$ measured from the center of the H. It is clear that, owing to the binding energy, $f(\boldsymbol{r})$ is appreciable only in the immediate vicinity of H, and spreads out as the temperature increases. At some moment the hole transfers to the spontaneous vortex, interchanging the roles of the S and the H. The practical effect of this transfer is that the hole moved to some neighbour plaquette at a new location $\boldsymbol{r}$. Since the position of the pivot has changed, the random rotation is now around some new centre. Later on, yet another such transfer takes place, affecting an additional translation of the hole and so on. Thus we end up with a mechanism that causes the complex to bootstrap itself with each transfer. This can be effectively described by a two-dimensional random walk where each transfer of the hole between the vortices can be viewed as a single jump with probability $f(\boldsymbol{r})$. This, however, is not a classical random walk process. We have already seen above that there is a typical probability that the hole will be found at the initial well. Therefore there is a typical time that the hole spends within the "pivot" well before moving to the other. This typical time τ is the mean of the waiting time distribution, $W(t)$. Evidently, this distribution is closely related to the probability of finding the hole within the vortex, $p_v(t)$. However, this probability, which has been calculated above, may differ from the present one. Nevertheless, just for demonstration, let us assume the same (normalized) distribution of waiting times,

$$W(t) = (4\,\Delta E/h)p_v(t)\,, \tag{8}$$

where $\Delta E \equiv (2\varepsilon\,\delta E)/(1-\varepsilon^2)$ is the energy difference between the symmetric and antisymmetric eigenstates. The mean and variance of this distribution are

$$\tau \equiv \langle t \rangle = \frac{h(1+\varepsilon^2)}{4\,\Delta E}\,, \tag{9a}$$

$$\langle \delta t^2 \rangle^{1/2} = \frac{h}{2\,\Delta E}\left(\frac{(1+\varepsilon^2)(1-2\varepsilon^2)}{12} + \frac{2(1-\varepsilon^2)}{\pi^2}\right). \tag{9b}$$

The values of both the mean and the variance are then proportional to $1/\Delta E \sim 1/\varepsilon$, which increases with the height of the barrier between the wells. Random walks with waiting time distributions were studied by Montroll and Weiss [10], who concluded that whenever the value of τ is finite, all the results for classical random walks apply when the number of steps (in two dimensions) is replaced by the rescaled time $(t/\tau)/\ln(t/\tau)$. The increase of the mean time with $1/\varepsilon$ indicates that the higher the barrier, the longer the hole spends in the well before jumping, and that one has to go to larger and larger times to obtain the asymptotic random walk behaviour.

This result can now be used for applying known properties of random walks to the system of dilute holes in high-temperature superconductors. As an example of such an application we derive the "mean free time" between the combination of two such SH excitations. Neglecting all possible interactions between the excitations (see discussion below), the dynamics of the hole transport is governed by the above mechanism alone. For simplicity we also assume that the holes are initially distributed uniformly over the plane with surface concentration $n = x/a^2$, where x is the doping concentration. The typical area that such a hole occupies is then $A = a^2/x$ (in general, as the temperature increases, the separation between the vortex and the antivortex grows as well, but this modification should not change the overall behaviour in the range of temperatures discussed here, and will only introduce a factor in the final result). The typical number of steps it takes for the complex to cover this space is then $N \approx A/2\lambda^2 \approx a^2/2x\lambda^2$, where λ is the typical length of each step and the factor of 2 is introduced to take account of the two translational degrees of freedom in the plane. Assuming that the S always rearranges as a nearest plaquette in one of the main lattice directions with respect to H, the length λ is then a. (However, numerical simulations [6] suggest that other configurations are also possible, e.g., mainly $\lambda = \sqrt{2}\, a$.) The typical mean free time to combine is then

$$T = N\tau = \frac{(a/\lambda)^2}{2x} \frac{h(1+\varepsilon^2)}{4\Delta E} . \tag{10}$$

Therefore the rate of combination is linked to the energy barrier between the vortex and the antivortex. The ratio $(a/\lambda)^2$ is a number of order one, and should not introduce any difficulty when trying to determine the parameters experimentally.

4. Discussion and conclusion

To conclude it has been shown that at low temperatures, a hole that leads to a SH excitation can be described as a particle in a double well potential. As long as higher excitations can be ignored, this leads to a time oscillation of the probability to find the hole between the vortex and the antivortex, which should be amenable to experimental verification by spectroscopic techniques. An assumption underlying the present analysis was that both rotation and translation of the complex of the hole in the vortex/antivortex excitation are very slow compared to the time of oscillation, and hence the DWP could be considered stationary in the calculation. In other words, we expect that the

above analysis holds as long as the typical time for "hopping" of the S is longer than $h/\Delta E$. As this condition is relaxed, the effective Hamiltonian of the DWP should be modified accordingly to include other degrees of freedom, namely, translational and angular momentum of the entire complex should be considered as well. Due to the simplicity of the Hamiltonian, it is interesting if a perturbative approach is feasible. It is evident that as the temperature increases, these higher degrees of freedom come into play and may change the picture. However, one should bear in mind that, irrespective of the temperature, as long as there exists a bound vortex/antivortex pair, there is a double degeneracy for the hole to occupy one or the other. Thus, intuitively, there must exist an oscillation of the probability to be found between the states and a resonance should be detected.

Another assumption involved neglecting the interaction with the rest of the system. In fact there are both Coulomb repulsion between the charges (decaying as $1/r$ since the charges are embedded in 3D) and a dipolar interaction between two SH complexes ($\sim 1/r^2$ since they are 2D entitites). This confines the validity of the analysis to low concentrations. To determine how low the concentration should be, one should find the distance, R_c, at which the magnitude of the interaction between two SH excitations is comparable to the height of the barrier between the wells. This defines some critical concentration $x_c \approx a/R_c$, and the dilute regime can be casted in the requirement $x < x_c$.

If the picture suggested here describes faithfully the low doping regime in high-temperature superconductors, then these materials are valuable for the study of basic questions that bear on the fundamental concepts of quantum mechanics. These questions relate, e.g., to *superselection* rules and parity symmetry breaking [9, 11, 12]. A more elaborate discussion of this ramification will be presented elsewhere [13].

Acknowledgements

It is a great pleasure to acknowledge critical and fruitful discussions with Prof. D.J. Thouless, Prof. N. Jan and Prof. A. Coniglio. This research is supported by a grant No. RG9358 of the Science and Engineering Research Council, UK.

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