Reply to "Validity of the t-J model"

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We respond to the preceding Comment of Zhang and Rice concerning a previous paper in which we questioned their claim to have derived an effective single-band model from a two-band model of high-temperature superconductors.

In a recent paper,[1] Emery and Reiter (ER) found an exact solution of a model for a mobile hole in the copper oxide planes of high-temperature superconductors. The hole moves on oxygen sites through a lattice of spins localized on copper, and the solution was found for a ferromagnetic background. We showed that, contrary to an assertion of Zhang and Rice[2] (ZR), the low-energy physics is not necessarily the same as for a single-band model.

The conclusion has been questioned by Zhang and Rice in the preceding Comment[3] (ZRC). Their argument is not persuasive: they mainly restate our solution for a special case, without accepting the full consequences, and criticize an approximation that we used for purposes of illustration. The latter criticism disproves a claim that we did not make and that, anyway, has no bearing on the validity of the t-J model.

We shall use the Hamiltonian for the model as defined in Refs. 1 and 2. It is characterized by the copper-copper exchange integral J and by effective hopping parameters t_1, and t_2 for an oxygen hole. The latter refer to intermediate states in which a Cu site is unoccupied (t_1) or doubly occupied (t_2). Although the physical exchange coupling is antiferromagnetic ($J>0$), ZR's argument did not assume a particular Cu spin configuration. Hence it is sufficient to study a ferromagnetic background as a counterexample. In ER, the problem was solved for arbitrary t_1 and t_2 in the conventional sense of giving explicit expressions for all eigenfunctions [Eqs. (6) and (9) of ER] in terms of an eigenvalue $\lambda_{\mathbf{k}}$ defined by an integral. Contrary to the statements in ZRC, our solution is not restricted to $t_1=0$, although we did show that, in that case, the integral may be carried out explicitly, so that it is rather easy to study the properties of the solution. Consequently we shall restrict our discussion to $t_1=0$ as in ZRC, although it suppresses the spread of the quasiparticle which may be important at higher carrier densities.

For $t_1=0$, the eigenvectors $|\psi^0_{\mathbf{k}}\rangle$ and eigenvalues $\lambda_{\mathbf{k}}$ are given by[1]

$$|\psi^0_{\mathbf{k}}\rangle=\left[a_{\mathbf{k}}b^{\dagger}_{\mathbf{k}\uparrow}-\frac{1}{N}\sum_{\mathbf{q}}a_{\mathbf{q}}b^{\dagger}_{\mathbf{q}\downarrow}S^{+}_{\mathbf{k}-\mathbf{q}}\right]|\mathrm{FM}\downarrow\rangle \tag{1}$$

and

$$\lambda_{\mathbf{k}}=-t_2(a^2_{\mathbf{k}}+4)\,, \tag{2}$$

where

$$a^2_{\mathbf{k}}=2(2+\cos k_x+\cos k_y)\,. \tag{3}$$

Here $|\mathrm{FM}\downarrow\rangle$ is the ferromagnetic state with all Cu spins $\downarrow$, $S^{+}_{\mathbf{k}-\mathbf{q}}$ is the Fourier transform of the spin raising operator, and

$$b_{\mathbf{k}\sigma}=(a_{\mathbf{k}}N^{1/2})^{-1}\sum_{\mathbf{m}+\Delta}a_{\mathbf{m}+\Delta,\sigma}e^{-i\mathbf{k}\cdot\mathbf{m}}\,. \tag{4}$$

Also, $a^{\dagger}_{\mathbf{m}+\Delta,\sigma}$ creates a hole of spin σ in an $O(2p_x)$ or $O(2p_y)$ state at $\mathbf{m}+\Delta$ where Δ is $(\pm\frac{1}{2},0)$ or $(0,\pm\frac{1}{2})$. Then, after a unitary transformation, the Hamiltonian may be written in the form

$$H=-t_2\sum_{\mathbf{k}}|\psi^0_{\mathbf{k}}\rangle\langle\psi^0_{\mathbf{k}}|\,. \tag{5}$$

Note that the $|\psi^0_{\mathbf{k}}\rangle$ are not normalized and the eigenvalues of H are given by Eq. (2) because $\langle\psi^0_{\mathbf{k}}|\psi^0_{\mathbf{k}}\rangle=a^2_{\mathbf{k}}+4$. The total number of states of spin $-(N-1)/2$ in the original *Hamiltonian* is $2(N+N^2)$. Of these, the N states $|\psi^0_{\mathbf{k}}\rangle$ form a band $\lambda_{\mathbf{k}}$ and the remainder contain nonbonding spatial or spin configurations and hence have energy equal to the oxygen site energy ε_p, which is taken to be zero.

ZRC notes that, from Eqs. (1) and (4), the $|\psi^0_{\mathbf{k}}\rangle$ may be rewritten as plane-wave superpositions of singlets formed by the Cu spin at $\mathbf{m}$ and the oxygen hole in a state created by

$$P^{\dagger}_{\mathbf{m},\sigma}=\tfrac{1}{2}\sum_{\Delta}a^{\dagger}_{\mathbf{m}+\Delta,\sigma}\,, \tag{6}$$

which is a sum over the four oxygen sites surrounding the Cu ion at $\mathbf{m}$. But this is *not* the Zhang-Rice singlet and it is *not* equivalent to a doubly occupied site in the single-band model.

The Zhang-Rice approximation uses singlets constructed from the Wannier states $b_{\mathbf{m}\sigma}$ which are the Fourier transforms of the $b_{\mathbf{k}\sigma}$. They are related to $P_{\mathbf{m}\sigma}$ by

$$P_{\mathbf{m}\sigma}=\sum_{\mathbf{n}}a(\mathbf{m}-\mathbf{n})b_{\mathbf{n}\sigma}\,, \tag{7}$$

where $a(\mathbf{m}-\mathbf{n})$ is the Fourier transform of $a_{\mathbf{k}}$. The $b_{\mathbf{n}\sigma}$ are used because they are orthogonal whereas the $P_{\mathbf{m}\sigma}$ are not. ZRC implies that the use of one or the other is a matter of convenience, but it is the central reason for the

difference between the copper-oxygen model and the t-J model. In order to get a clearer feeling for this difference, we introduce the Fourier transforms of in-cell singlets and triplets, involving the oxygen Wannier states

$$|\phi_{\mathbf{k}}^{\pm}\rangle = \frac{1}{\sqrt{2N}} \sum_{\mathbf{m}} e^{i\mathbf{k}\mathbf{m}} (b_{\mathbf{m}\uparrow}^{\dagger} \pm b_{\mathbf{m}\downarrow}^{\dagger} S_{\mathbf{m}}^{+}) |\mathrm{FM}\downarrow\rangle \tag{8}$$

$$= \frac{1}{\sqrt{2}} \left(b_{\mathbf{k}\uparrow}^{\dagger} \pm \frac{1}{N} \sum_{\mathbf{q}} b_{\mathbf{q}\downarrow}^{\dagger} S_{\mathbf{k}-\mathbf{q}}^{+} \right) |\mathrm{FM}\downarrow\rangle . \tag{9}$$

Now for $t_1 = 0$, the spread of the wave function is quite small[1] and it is a good approximation to write $|\psi_{\mathbf{k}}^{0}\rangle$ as a sum of $|\phi_{\mathbf{k}}^{+}\rangle$ and $|\phi_{\mathbf{k}}^{-}\rangle$. Using Eqs. (1), (8), and (9),

$$\langle \phi_{\mathbf{k}}^{\pm} | \psi_{\mathbf{k}}^{0} \rangle = \frac{1}{\sqrt{2}} [\alpha_{\mathbf{k}} \mp \alpha(0)] \delta_{\mathbf{k}\mathbf{k}'} \tag{10}$$

and then

$$|\psi_{\mathbf{k}}^{0}\rangle \approx [\alpha_{\mathbf{k}}^2 + \alpha^2(0)]^{1/2} [\cos\theta_{\mathbf{k}} |\phi_{\mathbf{k}}^{-}\rangle + \sin\theta_{\mathbf{k}} |\phi_{\mathbf{k}}^{+}\rangle] , \tag{11}$$

where the singlet-triplet mixing angle is given by

$$\tan\theta_{\mathbf{k}} = \frac{\alpha_{\mathbf{k}} - \alpha(0)}{\alpha_{\mathbf{k}} + \alpha(0)} . \tag{12}$$

The ZR approximation sets $\theta_{\mathbf{k}} = 0$ (pure singlet) but it is clear that $\tan\theta_{\mathbf{k}}$ ranges from essentially zero, when $\alpha_{\mathbf{k}} = 2$ to (-1) when $\alpha_{\mathbf{k}} = 0$. To illustrate the physical significance, let us calculate the z component of the oxygen spin-spin correlation function for the state $|\psi_{\mathbf{k}}^{0}\rangle$

$$S_{\mathbf{k}}^{z}(\mathbf{Q},\omega) \equiv \frac{1}{N} \sum_{\mathbf{k}'} \frac{|\langle \psi_{\mathbf{k}}^{0} | s_{\mathbf{Q}}^{z} | \psi_{\mathbf{k}'}^{0} \rangle|^2}{\langle \psi_{\mathbf{k}}^{0} | \psi_{\mathbf{k}}^{0} \rangle \langle \psi_{\mathbf{k}'}^{0} | \psi_{\mathbf{k}'}^{0} \rangle} \delta(\omega - \lambda_{\mathbf{k}} + \lambda_{\mathbf{k}'}) , \tag{13}$$

where $s_{\mathbf{Q}}^{z}$ is the Fourier transform of the z component of the oxygen spin operator $s_{\mathbf{Q}}^{z} \equiv \frac{1}{2} \sum_{\mathbf{p}\sigma} \sigma b_{\mathbf{p}+\mathbf{Q},\sigma}^{\dagger} b_{\mathbf{p}\sigma}$ [omitting the nonbonding states which will not contribute to Eq. (13)]. It is easy to evaluate $S_{\mathbf{k}}^{z}(\mathbf{Q},\omega)$ using the exact eigenfunctions but it is more instructive to use the approximation (11) to find

$$S_{\mathbf{k}}^{z}(\mathbf{Q},\omega) = \frac{1}{4N} \sin^2(\theta_{\mathbf{k}} + \theta_{\mathbf{k}+\mathbf{Q}}) \delta(\omega - \lambda_{\mathbf{k}} + \lambda_{\mathbf{k}+\mathbf{Q}}) . \tag{14}$$

For many oxygen holes the result is intensive because $S_{\mathbf{k}}^{z}(\mathbf{Q},\omega)$ should be summed over k values in a Fermi sea (assuming independent quasiparticles). In principle, $S_{\mathbf{k}}^{z}(\mathbf{Q},\omega)$ is a measurable quantity, but it is zero in the ZR approximation ($\theta_{\mathbf{k}} = 0$, all $\mathbf{k}$). Indeed they set all oxygen spin-spin correlation functions equal to zero—a result that has been used in the interpretation of NMR experiments.[4] However, it is evident that the spin reduction factor $|\sin(\theta_{\mathbf{k}} + \theta_{\mathbf{k}+\mathbf{Q}})|$ is not small in general—indeed it is equal to 1 when $\mathbf{k} = (\pi,\pi)$ and $\mathbf{Q} = 0$. Moreover $S_{\mathbf{k}}^{z}(\mathbf{Q},\omega)$ clearly is part of the low-energy physics since the energies in the δ functions are in the low-energy band.

ZR's argument was that there is a large gap between the singlet and triplet states so the triplets may be neglected. For the present problem, the singlet-triplet splitting is $2t_2\alpha_{\mathbf{k}}\alpha(0)$ which actually vanishes at $\mathbf{k} = (\pi,\pi)$. But, even where it is not small, there is a significant mixing matrix element $+t_2[\alpha^2(0) - \alpha_{\mathbf{k}}^2]/2$. As pointed out in ER, the overlap $\cos\theta_{\mathbf{k}}$ between $|\psi_{\mathbf{k}}^{0}\rangle$ and the singlet state is not necessarily a good measure of the mixing—it is better to look at the triplet amplitude $\sin\theta_{\mathbf{k}}$. (The value of $\cos\theta_{\mathbf{k}}$ was given in ER and plotted in Fig. 1 of ZRC.) For example, as pointed out in ER, when $\mathbf{k} = 0$, $\mathbf{Q} = 0$, $\cos\theta_{\mathbf{k}} = 0.98$ [from Eq. (12)] but the spin reduction factor $\sin 2\theta_{\mathbf{k}} = 0.37$.

ZRC's only comment about all of this is to point out that the spin on oxygen is not a good quantum number. This is true and, by the same token, the total spin on copper is not a good quantum number (since the overall spin *is* conserved). Thus ZRC's remark does not contradict our conclusion but reinforces it, since the spin on copper *is* conserved in the t-J model.

We now turn to ZRC's criticism of an approximation that we used for purposes of illustration.

ZRC states "(ER) propose that the correct basis to describe the result is composed of a hole on a single O atom coupled to its two neighboring Cu atoms, i.e., basis states of the form $6^{-1/2}[2|\downarrow\uparrow\downarrow\rangle - |\uparrow\downarrow\downarrow\rangle - |\downarrow\downarrow\uparrow\rangle]$." This is incorrect and the comment in ZRC that this is not a good basis *for all* $\mathbf{k}$ has nothing to do with what was said in ER or the validity of the t-J model. Obviously our *basis* is the orthogonal set of states $|\psi_{\mathbf{k}}\rangle$ which diagonalize H when $t_1 = 0$. What we actually said was that, *for small* k, it is useful to think of the oxygen hole hopping between sites with the Cu-O-Cu spin configuration mentioned above. This statement is correct, and is not disproved by showing that the approximation is not good for $\mathbf{k} = (\pi,\pi)$ as claimed in ZRC. The representation was used mainly to illustrate why there is a spin associated with the charge and is unrelated to the validity of the t-J model since that involves ZR's approximation, not ours. It is straightforward to show that for small k the overlap between the exact eigenfunctions and a plane wave made up of our states is $(1 - k^2/48)$. Moreover, when k is small the average spin on the oxygen site is $\frac{1}{3}(1 - k^2/6)$ for the exact eigenfunction and $\frac{1}{3}$ for our approximation. Therefore, we believe that the approximation is good *for small* k as claimed.

Next we should like to comment on three papers cited by ZRC in support of their contention.

(i) Mila[5] and separately Eskes and Sawatzky[6] found a local Cu-O singlet in a model with *only one copper ion* surrounded by oxygen ions. We, too, would find a local Cu-O singlet for that case, but it obviously has no bearing on the issues under discussion, since hopping of the oxygen hole from the neighborhood of one copper site to another is essential for the singlet-triplet mixing.

(ii) Schüttler and Fedro[7] have carried out numerical calculations for a periodically extended system of four Cu and eight O sites containing five or six holes. They concluded that their results were inconsistent with our picture. However, we believe that if they calculated $S_{\mathbf{k}}^{z}(\mathbf{Q},\omega)$ for a range of values of $\mathbf{k}$ and $\mathbf{Q}$, the differences from the t-J model would become clear.

(iii) Zhang[8] has considered a single oxygen hole for the case $t_1 = 0$, and no Cu-Cu superexchange. He shows that, for an arbitrary background spin state, there is a correspondence between all the states of the t-J model and the lower band for the oxygen hole. Specifically, the ener-

gy spectra and number of degrees of freedom are the same and the wave functions are formally related by replacing a doubly occupied site of the t-J model by a Cu-O singlet involving the nonorthogonal states $P^{\dagger}_{\mathbf{m},\sigma}$ defined in Eq. (6). Zhang claims that this result shows that the two models have the same low-energy physics. We disagree. The difference in the wave functions surely is important. It would give rise to a nonvanishing oxygen spin-spin correlation function as we have shown here.

Finally we should like to add that a clear distinction between the models can be seen when the on-site Coulomb interactions are large. They are denoted by U in the single-band model and by U_p or U_d for O($2p$) and Cu($3d$) states in the copper-oxygen model. ZR omitted U_p, and their calculation would be significantly modified if it were included. However, it seems to be a strong interaction in high-temperature superconductors—almost as large as U_d. For one hole per unit cell, the single-band model is dead in the $U \rightarrow \infty$ limit: there is no hopping, no exchange, and the ground state for N sites is 2^N-fold degenerate. By contrast, when U_p and $U_d \rightarrow \infty$, the copper oxygen model still has a four-spin exchange and the ground state has antiferromagnetic order.[9] Moreover, even if the four-spin exchange is omitted, a mobile hole favors a ferromagnetic state in the single-band model,[10] but favors a low-spin state in the copper oxygen model.[11]

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