On the nature of high-temperature superconductivity

by V. J. Emery

A picture of the electronic structure, magnetism, and superconductivity in high-T_c oxides is obtained from a simple analysis of experiments and models of the copper oxide planes. It is shown that magnetism is associated with holes on copper and superconductivity with holes on oxygen. The pairing force is not retarded. Questions about the motion of charges in an antiferromagnetic background and the many-body theory of high-temperature superconductivity are discussed. Differences between the cuprates and doped $BaBiO_3$ are emphasized.

1. Introduction

Despite two years of intensive effort since the first announcement of high-temperature superconductors [1], there is no real agreement about how the properties of these fascinating materials are to be explained. It seems clear that the simple free-electron picture is inadequate, that a new mechanism of superconductivity is required, and that significant developments of many-body theory will have to be made. The challenge is to uncover the essential features of these complicated, multicomponent systems, and to replace the conceptual fabric that has served us so well for conventional superconductors.

In such a situation, it is useful to take a rather simple point of view and to seek out the critical experiments that are most likely to point us in the right direction. Although the selection of what is important is to some degree a matter of taste, such an exercise may well give a good indication of where further effort is most useful. Here we shall update and expand a discussion of this sort that was published somewhat more than a year ago [2]. The conclusions reached there and in an earlier paper [3] continue to hold good: The magnetic behavior of high-temperature superconductors is associated with holes largely on copper sites; superconductivity is produced by holes on oxygen, and pairing is caused by a high-energy, nonretarded interaction.

2. Electronic structure and magnetism

The essential properties of the electronic structure of the copper oxide planes, the common feature of almost all high-temperature superconductors, may be obtained by valence counting and simple energy considerations [3].

Consider first $La_{2-x}Sr_xCuO_4$. Starting from the closed-shell configurations La^{3+}, Sr^{2+}, Cu^+, and O^-, charge balance requires that the average number of holes available for the CuO_2 planes be $1 + x$ per formula unit. Suppose now that the energies to add a hole into the Cu(3d) and O(2p) states are denoted by ε_d and ε_p, respectively. Let the Coulomb interactions between holes be U_d on copper, U_p on oxygen, and V for neighboring copper and oxygen sites. These quantities are estimated to be $\varepsilon \equiv \varepsilon_p - \varepsilon_d \approx 1$–$2$ eV; $U_d \approx 8$–10 eV; $U_p \approx 5$–7 eV; and $V \approx 1$–2 eV. Then, in the absence of hopping, the first holes will go into the lowest-energy Cu($3d$) states to produce Cu^{2+}. This is the situation in La_2CuO_4. However, additional holes produced by doping with Sr will go into O($2p$) states because their energy $\varepsilon + 2V$ is smaller than U_d. When hopping due to overlap of orbitals is taken into account, the relatively low density of O($2p$) holes may delocalize over the entire system to form a band, but the strong Coulomb interaction still enforces localization of the Cu($3d$) holes. [We shall continue to refer to "O($2p$) holes" and "Cu($3d$) holes," although there is of course some mixing of the two.] It is well known from the theory of the one-dimensional electron gas [4, 5] that the near-neighbor Coulomb interaction V is essential for the charge-ordering of the Cu($3d$) holes at such a low density. Hopping also induces an antiferromagnetic superexchange interaction between the Cu^{2+} spins and gives rise to the magnetic ordering and dynamical correlations observed in all of the cuprate superconductors [6(a)]. This is a kinetic effect: A hole has a lower zero-point energy when it is surrounded by opposite spins, since it may then make excursions onto neighboring sites without violating the exclusion principle.

Putting this all together, it can be seen that magnetism is associated with holes on copper and superconductivity with the very low concentration of mobile holes on oxygen.

A similar but slightly more complicated argument may be made for $YBa_2Cu_3O_{6+y}$. Here it is necessary to make use of the additional information that some of the copper atoms in the CuO chains remain in the Cu^{+} state [7]. In the tetragonal phase the copper configurations are $Cu^{+}_{1-2y}Cu^{2+}_{2+2y}$, and charge is balanced if the remaining ions are Y^{3+}, Ba^{2+}, and O^{2-}. There are no excess oxygen holes, and the material is antiferromagnetic [6(a)]. On the other hand, the copper configuration in the orthorhombic phase is $Cu^{+}_{1-y}Cu^{2+}_{2+y}$, and there are y oxygen holes per formula unit to divide between the oxygen atoms on the CuO chains and CuO_2 planes. This explains why superconductivity is associated with the orthorhombic phase, with carrier concentration that increases with oxygen content.

It has steadily become clear that this picture of the electronic structure is appropriate for the CuO_2 planes in all of the cuprate superconductors, although simple valence-counting arguments cannot be used to obtain the available charge in the less ionic Bi and Tl compounds. Overwhelming support can be found in a wide variety of spectroscopic measurements and detailed studies of magnetic properties using neutron-scattering and magnetic resonance techniques. In particular, X-ray absorption near-edge spectroscopy [8] shows that the copper remains in the Cu^{2+} state for Sr concentrations up to at least $x = 0.3$, where $La_{2-x}Sr_xCuO_4$ once again becomes an insulator. Furthermore, the antiferromagnetic order parameter and form factor are very close to what is expected for a two-dimensional array of Cu^{2+} ions [9]. The most direct evidence for oxygen holes comes from the use of electron energy-loss spectroscopy [6(b)] to study the excitation of electrons from $1s$ to $2p$ states. This is forbidden in O^{2-} because the O($2p$) states are already occupied, and indeed it is not seen in La_2CuO_4. However, it appears when Sr is added.

More detailed descriptions of the experiments and references to the original papers may be found in companion papers in this issue [6(a)–(e)].

A major question remaining is whether the O($2p$) holes that are responsible for superconductivity have $p\sigma$ or $p\pi$ character. Current experiments [10] rule out p_z orbitals, but so far have not been able to provide a clear distinction between the in-plane states. Band structure calculations [11] suggest that $p\sigma$ are the relevant states, but small cluster studies [6(a), 12] favor $p\pi$. The physical reason for this is quite clear: The $p\pi$ states minimize the Coulomb interaction with the holes on Cu, whereas $p\sigma$ states have a better overlap with the Cu orbitals and therefore a lower kinetic energy.

3. Low-energy physics

The Hamiltonian for the model described in Section 2 may be written

$$H = \sum_{\substack{\langle \mathbf{i},\mathbf{j}\rangle \\ \sigma}} \varepsilon_{\mathbf{ij}} a^{\dagger}_{\mathbf{i}\sigma} a_{\mathbf{j}\sigma} + \frac{1}{2} \sum_{\mathbf{i},\mathbf{j}} U_{\mathbf{ij}} n_{\mathbf{i}} n_{\mathbf{j}}, \tag{1}$$

where $\mathbf{i}$ is (m, n) for a Cu site and $(m + 1/2, n)$ for an oxygen site. The $a^{\dagger}_{\mathbf{i}\sigma}$ create holes of spin σ at site $\mathbf{i}$, and it is assumed that a factor $(-1)^{m+n}$ is absorbed into the $a^{\dagger}_{\mathbf{i}\sigma}$ to take account of signs related to the symmetry of the Cu($3d$) states. Also, $n_{\mathbf{i}} \equiv a^{\dagger}_{\mathbf{i}\uparrow}a_{\mathbf{i}\uparrow} + a^{\dagger}_{\mathbf{i}\downarrow}a_{\mathbf{i}\downarrow}$ is the number operator at site $\mathbf{i}$. In terms of the notation of Section 2, the site-diagonal terms $(\varepsilon_{\mathbf{ii}}, U_{\mathbf{ii}})$ are given by (ε_p, U_p) and (ε_d, U_d) for the O($2p$) and Cu($3d$) states, respectively, and $U_{\mathbf{ij}} = V$ for the neighboring Cu–O sites. If we include a hopping $\varepsilon_{\mathbf{ij}} = t$ between CuO neighbors, we obtain the model introduced in [3], but it may also be desirable to add a direct hopping between oxygen sites.

For weak hopping, the problem may be simplified by carrying out an expansion in powers of t, thereby removing some degrees of freedom and eliminating almost all of the strong interactions. For $t = 0$, the ground state described in Section 2 is degenerate because the energy does not depend on the spin of the $3d$ holes or the spin or position of the $2p$ holes. Degenerate perturbation theory in powers of t leads to an effective Hamiltonian H_{eff}, acting within the degenerate subspace and containing the following terms [3, 13]:

1. An effective O($2p$)–O($2p$) hopping of O(t^2), with or without spin flip.
2. A superexchange interaction between neighboring O($2p$) and Cu($3d$) spins with exchange constant

$$J = 2t^2 \left[\frac{1}{\varepsilon + U_p - V} + \frac{1}{U_d - 2V - \varepsilon} \right]. \tag{2}$$

3. A superexchange interaction between neighboring Cu($3d$) spins with exchange constant

$$J_c = \frac{4t^4}{(\varepsilon + V)^2}\left[\frac{1}{U_d} + \frac{2}{U_p + 2\varepsilon}\right]. \tag{3}$$

4. An effective attractive interaction between O(2p) holes separated by one or two copper sites, mediated by the spins or zero-point fluctuations of the Cu(3d) holes.
5. The bare Coulomb interaction U_p.

A detailed discussion of the effective interaction is given in [13]. H_{eff} constitutes an effective low-energy Hamiltonian because all of the original strong interactions, except for U_p, have been eliminated. It is not difficult to deal with a large U_p when the density of oxygen holes is so low.

Naively, one might treat the O(2p) holes as a dilute Fermi gas, and indeed both photoemission [14] and position annihilation [6(f)] experiments do show evidence of a Fermi surface. However, in order to establish that such an approach is reasonable, it is necessary to get a better understanding of how the mobile holes destroy the long-range antiferromagnetic order of the Cu^{2+} spins, what is the nature of the residual magnetic correlations, and whether it is possible to have quasiparticles with a well-defined spin and charge. These are complicated many-body problems that must be addressed in any model of high-temperature superconductors.

An alternative approach [15] that is widely followed is to use a single-band Hubbard model instead of one involving both copper and oxygen sites. The idea is to diagonalize the copper–oxygen hopping Hamiltonian and to retain only states in the partially filled band. The Hamiltonian may be written in the form (1) if $a^\dagger_{\mathbf{i}\sigma}$ is reinterpreted as a creation operator for the corresponding Wannier state of cell **i**. In the simplest versions of the model, there is only a hopping $\bar{t}$ between neighboring cells and an on-site interaction U. Following the same argument as before, it is possible to derive an effective low-energy Hamiltonian in the strong-coupling limit. When $\bar{t} = 0$, the lowest-energy state for $U > 0$ has the minimum concentration of doubly occupied sites, i.e., δ of the hole concentration $1 + \delta$. The ground state is degenerate because its energy is independent of the spin of the singly occupied sites and the location of the doubly occupied ones. For small $\bar{t}$ the degeneracy is broken by the hopping $\bar{t}$ of one of the two holes on a site and by a superexchange interaction $J = 4\bar{t}^2/U$ between spins. Transforming from holes to electrons, we obtain the so-called t–J model [16] of mobile vacancies (missing spins) in an antiferromagnet. Recent work on this model has shown how an antiferromagnetic background affects the motion of a *single* vacancy [17], but there is some way to go in understanding the reverse problem of the influence of a *finite concentration* of vacancies on the magnetic state itself.

It is important to explore the differences between the single-band and copper–oxygen models. An obvious one can be seen by considering the consequences of *removing* holes from the $Cu^{2+}O_2^{2-}$ planes. For the single-band model, this is equivalent to adding holes, because it is symmetric under a particle–hole transformation. For the copper–oxygen model, it is quite different because the holes are removed from the copper sites to give Cu^+. The low-energy Hamiltonian is essentially the t–J model, and we have to contrast vacancies on copper with holes on oxygen, each moving through an antiferromagnetic background. Zhang and Rice [18] have argued that mathematically the two are equivalent, because a hole on oxygen forms a singlet state with the hole on the copper site of the same cell, and that singlet behaves in the same way as a doubly occupied site in the single-band model. This has been contested on the grounds that, correctly constructed, the singlets carry a spin as well as a charge, so their quantum numbers are different from those of a doubly occupied site [19]. Moreover, in the U, U_p, $U_d \to \infty$ limit, a vacancy in the single-band model enforces a ferromagnetic state of the background spins, whereas an oxygen hole prefers antiferromagnetic correlations [20]. In the same limit, the copper–oxygen model has a four-spin exchange [21] which produces a ground state with antiferromagnetic order, but the single-band model reduces to noninteracting spins.

It is sometimes suggested that all of this has little to do with superconductivity or the nature of the superconducting state. With increased doping or oxygen content the Cu(3d) and O(2p) holes would merge into a single Fermi liquid, exhibiting no strong magnetic effects, and superconductivity would have some other origin such as charge polarization. However, this point of view is difficult to sustain. Magnetic correlations have been observed [22] in a single crystal of $La_{2-x}Sr_xCuO_4$ which has $x = 0.11$ and is superconducting below 10 K. Moreover, Raman scattering [23] shows quite broad magnon peaks in $YBa_2Cu_3O_7$. However, it is equally important to note that the electronic structure which gives rise to magnetic effects remains intact: Spectroscopic evidence for Cu^{2+} and excess holes on oxygen sites extends all the way to materials with the highest T_c. The density of charge carriers continues to be given by the excess (oxygen) holes, a property that is closely related to the existence of localized Cu holes and the gap in the spectrum for charge excitations. Thus, it is easier to believe that the local moments on Cu persist, but are more difficult to observe as the magnetic correlation length decreases and dynamical properties are modified by the mobile holes. Certainly any alternative explanation must come to terms with the implications of the high-energy spectroscopy, which so far it has proven to be consistent with low-energy experiments, where they have been performed.

4. Superconductivity

According to the BCS theory [24, 25], superconductivity is a consequence of an instability of the Fermi sea leading to the formation of (Cooper) pairs of electrons. The pairs form because there is an attractive interaction due to exchange of phonons which is sufficiently retarded to overcome the Coulomb repulsion.

It has been clear for some time that there are pairs in the high-T_c superconductors [26]. But the coherence length, the size of a Cooper pair, is exceptionally short [6(g)]—about 10 Å in $YBa_2Cu_3O_7$, which is less than twice the average distance between the oxygen holes. This has led to speculations that it is better to think of pre-existing or "real-space" pairs formed at some high temperature rather than, in the BCS way, at T_c. In this alternative picture, superconductivity would simply be a consequence of Bose condensation, a macroscopic occupation of the zero-momentum state for center of mass motion of the pairs. Something similar happens in liquid He^4. The helium atoms become bound states of alpha particles and electrons at temperatures of several million degrees, but it is only at about 2 K that their center of mass motion Bose-condenses to form a superfluid. It is usually argued that real-space pairing in a metal is prevented by the Coulomb interaction, which must be very strong because the size of such a pair is necessarily much smaller than the average spacing between electrons. But in high-T_c superconductors, Cooper pairs must face the same problem because the coherence length is so short and, as we shall see, the pairing force is not retarded.

A more significant difficulty is that the transition temperature for real-space pairing seems to be too high. A Bose gas in two dimensions does not condense but would begin to have a significant diamagnetism at a temperature $T_0 = 2\pi n\hbar^2/m$, where n is the areal density of the pairs and m their effective mass. The value of T_0 may be estimated from the low-temperature value of the penetration depth which, in the London limit that is appropriate for high-T_c superconductors, is given by

$$\lambda^{-2} = \omega_p^2/c^2 = 4\pi ne^2/mc^2d, \tag{4}$$

where ω_p is the plasma frequency, c is the velocity of light, and d is the average spacing between CuO_2 planes. For $YBa_2Cu_3O_7$, λ is about 1500 Å [27, 28] and for $d = 6.5$ Å, T_0 is about 4500 K. The actual ordering temperature will be lower because of interactions and the need to establish full three-dimensional phase coherence, but these effects are unlikely to give a factor of 50. An alternative possibility is that the reduction is brought about by strong inelastic scattering of the charge carriers [29], but this has yet to be explored in detail.

At the same time, the short coherence length need not be a severe problem for BCS theory. A better criterion is that the energy gap Δ is much smaller than the Fermi energy. The latter is given by $p_F^2/2m^*$ for a quadratic band with effective mass m^* and Fermi momentum p_F, and may be obtained from the value of the penetration depth for fermions [$m \rightarrow m^*$ and $n \rightarrow p_F^2/2\pi\hbar^2$ in Equation (4)]. Estimating Δ from T_c, we find that $2m^*\Delta/p_F^2$ is about 0.05, which is much larger than for low-temperature superconductors but nevertheless is small enough for the general ideas of BCS theory to be valid. Indeed, an expansion in powers of $2m^*\Delta/p_F^2$ should be adequate for developing a systematic theory. To this should be added the caveat that some experiments give larger values of m^* and Δ than were used in this estimate, and the argument would be less persuasive if they turned out to be correct.

On the experimental side, there is some evidence for both pictures. It has been argued [30] that the transport properties require bosons in the normal state. On the other hand, the observation of a Knight shift [31] and a Hebel–Schlichter peak in the nuclear spin relaxation rate [6(c), (d)] both favor the BCS picture.

We now turn to the pairing force itself and to what is perhaps the most significant constraint on the possible mechanism of high-temperature superconductivity—that the pairing force is nonretarded [2]. This is just the opposite of a phonon-mediated interaction where the lattice is slow to relax, so two electrons are able to feel its influence at different times, thereby avoiding the Coulomb repulsion. By the uncertainty principle, an equivalent way of saying the same thing is that the phonon energy scale (10^2 K) is much smaller than the electronic energy scale (10^4 K). This is why in BCS theory T_c is proportional to the Debye temperature θ_D, which is the range of energies over which the electron–phonon interaction is effective. In the other (nonretarded) extreme, where the energy of the excitations that are responsible for pairing is high [32], T_c is proportional to $p_F^2/2m^*$. This limit is most easily realized in low-density systems such as atomic nuclei or, potentially, helium mixtures where p_F is small.

The first evidence that the interaction is not retarded [2] came from the Hall effect, which indicated that T_c is proportional to n. Subsequently a systematic study of the penetration depth λ was undertaken [27] in a range of samples of $La_{2-x}Sr_xCuO_4$, $YBa_2Cu_3O_{6+y}$, and the Tl- and Bi-based materials. It was found that T_c is proportional to λ^{-2} all the way up to $T_c = 125$ K. This is confirmed by the observation [33] that T_c is proportional to ω_p^2, and according to the discussion below Equation (4), both imply that T_c is proportional to $p_F^2/2m^*$ and hence that the interaction is not retarded. Notice also that real-space pairing gives the same result. The same experiments indicated that $p_F^2/2m^*$ is about 0.5 eV for the three-plane Tl materials, so the energy scale of the excitations that mediate the pairing force should exceed this value. Phonons will not do, and even the high magnetic energy scale $2J \approx 0.24$ eV is too low. Only excitations that involve electronic intermediate states can work.

This does not necessarily mean that in a BCS context the pairing mechanism must be related exclusively to the polarization of charge. Indeed, that very language is tailored to the customary weak-coupling perturbation view of a many-body system; but it is inappropriate for the high-temperature superconductors. When a low-energy Hamiltonian is derived by a weak hopping expansion, the induced interactions are kinetic in origin and should not be regarded as a "polarization" effect. Superexchange is an example. As we have seen, it reflects a decrease in zero-point kinetic energy when two holes of opposite spin are on neighboring sites. For this process, the intermediate states are electronic excitations that are compatible with the absence of retardation. This is not so at longer distances, since exchange of spin fluctuations involves some low-energy intermediate states where only spins are excited [13]. Thus it may be concluded [13] that a magnetic mechanism of high-temperature superconductivity must involve interactions between neighboring spins if it is to be consistent with the variation of T_c with $p_F^2/2m^*$.

How then is the attraction able to overcome the strong short-range repulsion without the aid of retardation? The answer lies in the low density of oxygen holes. Scattering at low-relative-momentum p_F does not probe the strong short-range repulsion and allows the attractive interaction to be effective [32]. The low relative momentum also favors *s*-state pairing [13], as indicated by the temperature dependence of the penetration depth measured by muon-spin rotation [28] and more recently by magnetization measurements on single crystals [34].

The effective attractive interaction in the low-energy Hamiltonian for the copper–oxygen model, described above, is able to account for high-temperature superconductivity with these properties, provided the Coulomb interaction V is large enough. A detailed account is given in [13].

5. Doped $BaBiO_3$

We conclude with some remarks on the differences between the cuprates and doped $BaBiO_3$. The lead-doped material $BaBi_xPb_{1-x}O_3$ was extensively studied about ten years ago [6(h)]. More recently, a T_c of almost 30 K has been obtained by doping with potassium onto the Ba sites [6(h)]. Since these materials do not contain copper and so far have not displayed any striking magnetic properties [35], it is natural to look for a common mechanism of superconductivity in oxides that does not involve magnetism. However, it is clear that at least the electronic structure of doped $BaBiO_3$ is rather different from that of the cuprates. This stems from the fact [36] that the energy ε_s to put a hole in the Bi(6*s*) state of Bi^{3+} is more than 2 eV *higher* than ε_p. Thus, in an ionic picture, the holes are on oxygen even in the insulating $BaBiO_3$. Instead of going into a small Cu(3*d*) state, a hole is spread around the six oxygen sites surrounding a Bi ion. Clearly it does not cost a large energy U to add a second hole into a cell, because the probability of having a doubly occupied oxygen site is quite small. Thus, an itinerant picture is more appropriate, and localized antiferromagnetism is not to be expected. Actually, magnetic order in $BaBiO_3$ is forestalled by a breathing-mode lattice distortion and associated charge-density wave, sometimes described as a disproportionation of Bi^{4+} into Bi^{3+} and Bi^{5+} This terminology should not be taken too literally. Even allowing for Bi–O hybridization, most of the amplitude of the hole wave function resides on oxygen sites, and it is better to talk of a charge-density wave on oxygen. When some Ba^{2+} is replaced by K^+, more holes are added to the oxygen sites, destroying nesting of the Fermi surface, removing the charge-density order, and allowing the material to become conducting.

In these circumstances, it is reasonable to suppose that superconductivity is a consequence of interaction between the holes and the coupled phonons and charge-density waves [6(h)]. However, there is no *a priori* reason to believe that the same should be true of the cuprates. The common element is that holes on oxygen are responsible for superconductivity in both cases. In other respects, though, the electronic structure is very different. Nature may have been kind enough to give us two novel mechanisms of superconductivity in the high-T_c oxides.

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