Electron-phonon interactions in quantum percolation theory

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The strength of electron-phonon coupling in layered high-temperature superconductors as described by quantum percolation theory can be larger than in ballistic metals as described by BCS theory, but only if the percolation is preceded by local superconductivity. Experimental evidence for such electronic precursors in Y-Ba-Cu-O has recently been observed in ion-channeling experiments.

I. INTRODUCTION

Layered cuprate high-temperature superconductors (HTSC) exhibit many anomalous properties, such as superconductive transition temperatures $T_c \geq 100$ K and normal-state resistivities linear in T. All of these anomalies are consistently explained[1-3] within the general framework of quantum percolation theory (QPT). Exotic or nonphonon theories of superconductivity begin with reduced Hamiltonians and attempt to explain the anomalies of HTSC in terms of novel interactions.[4] By contrast, QPT utilizes the same reduced Hamiltonian containing electron-phonon interactions as the conventional BCS theory of superconductive metals ($T_c \leq 25$ K, constant normal-state resistivities at low T), but it focuses on the nature of the electrically conductive states near a metal-semiconductor transition. In this way it is able to relate the normal-state resistivity anomaly to spectroscopic anomalies in the superconductive state[3] and also to explain why T_c is so large in terms of anomalously strong electron-phonon interactions at Fermi-energy-pinning interlayer defect states.[5]

At present no Hamiltonian theory is known that is capable of rigorously deducing the nature of the metal-superconductor transition, for instance in Si:P impurity bands.[6] The reason for this is clear: the transition is essentially percolative in nature, and even scaling theories are inadequate even for the simpler case of classical percolative transitions. However, a set-theoretic or topological approach[6] to disorder near the transitions has succeeded remarkably well in explaining all the salient features of the Si:P data, without or with compensation.[6] It is this set-theoretic approach that underlies QPT.

The set-theoretic approach has not gained general acceptance.[7,8] This is not surprising, because nonperturbative, non-Hamiltonian theories are rare in theoretical physics—about as rare, in fact, as HTSC is in materials science. However, we have already a number of examples[2,3,9] showing specific successes of QPT in explaining HTSC. In this paper I extend my earlier work to show how the set-theoretic approach, previously used to classify electronic states, can also be used to classify phonon states and electron-phonon interactions in HTSC. In this way the similarities and differences between QPT and Hamiltonian models are brought out more clearly, and the advantages of QPT in describing microscopically inhomogeneous materials become more apparent.

II. QPT CLASSIFICATION OF ELECTRONIC STATES

The central idea in QPT is that near certain kinds of metal-semiconductor transitions it is possible to separate electronic states near E_F into two sets, localized and extended. This separation need not be exact; indeed it may only be asymptotic. The prototypical example[6] is Si:P, where the density of extended states $n_e(E)$ is proportional to $(E-E_c)^{1/2}$ and ideally at the critical concentration of P impurities $n=n_c$, $E_F=E_c$. It is possible that this expression for $n_e(E)$ is valid only in a limited range of energies $E_c < E \ll E_c + E_I$, where E_I is the impurity binding energy.

The possibility of the separation of $n(E)=n_e(E)+n_l(E)$ into localized and extended components in Si:P is atypical. With more than 10–20 % compensation of the P donors by trivalent acceptors, the separation fails, according to the theory because all the P impurities are no longer equivalent because of acceptor local fields. Also the separation requires dimensionality $d>2$ because for $d \leq 2$ the states all form localized domains, much as in the random-field Ising model.[6,10] This dependence on dimensionality d actually is crucial for the layered cuprates, in which the disorder is certainly much less ideal than in Si:P. However, in first approximation $d=2$ for the electronic cuprate conductive layers, which are separated by semiconductive layers such as LaO_2 in $(La,Sr)_2CuO_4$ or BaO in $YBa_2Cu_3O_7$. According to QPT, for these materials to be conductive the dimensionality d must be $2+\epsilon$, with $\epsilon>0$. This is accomplished by interlayer defect bridges,[5] for instance Sr in $(La,Sr)_2CuO_4$ or apical oxygen vacancies in $YBa_2Cu_3O_7$. These interlayer bridges are easily counted (like the P donors in Si:P) and they also contribute to $n(E)$ and $n_e(E)$ for E near E_F. In their absence $n_e(E)=0$.

When we separate $n(E)$ for E near E_F into $n_e(E)$ and $n_l(E)$, we have a kind of two-fluid model,[6] in contrast to conventional Hamiltonian theories where the presence of disorder is usually treated by one-component Fermi-liquid theory. The two approaches lead to qualitatively

different predictions for normal-state behavior. One-component Fermi-liquid theory predicts,[11] from the normal-state resistivity which is linear in T, a specific heat γ that diverges logarithmically at $T\rightarrow 0$. My two-component theory predicts[3] a constant γ; in other words, the resistivity anomaly arises from a selection rule on scattering of normal states by residual disorder. This scattering produces no thermal anomalies. When sufficiently large quantities of $Bi_2Sr_2CuO_6$ (or some similar compound) with $T_c \lesssim 10$ K become available to permit specific-heat measurements, it should be possible to distinguish the two theories.

It appears that QPT has already succeeded in explaining[3] the changes in the electronic continuum in Y-Ba-Cu-O from above T_c to below T_c, from a constant scattering strength to a scattering strength linear in $|E-E_F|$. Thus we can say that there is tangible spectroscopic evidence for the separability of $n_l(E)$ and $n_e(E)$. This separation is also consistent with more complete tunneling data.[3,9]

III. PHONON STATES AND ELECTRON-PHONON COUPLING

While the electronic states are localized (apart from dopants and defects), the vibrational states are much less localized. Because of mass differences normal modes which are cation-centered may be localized to some extent in layers but normal modes based on the anion oxygen sublattice must be extended and phase coherent between layers, apart from a small number of modes with $\mathbf{k}$ very near symmetry points. As a first approximation this leads to the classification of all phonon modes as extended.

Before proceeding to the next step we must recognize that for HTSC we are in the strong-coupling limit[12] $\lambda \gtrsim 2$. In this limit all electronic quasiparticles are polarons, composite electron-phonon complexes. The following selection rules appear, however, to remain valid even in the strong-coupling limit.

The basic observation now is that the characters of products of states are quite simple: extended times extended equals extended, and extended times local equals local. Thus phonons scatter extended states only among themselves, and local states only among themselves.[13] This means that so far as the extended states are concerned, even though they are not Bloch states, but are instead complex states which percolate from layer to layer in a phase-coherent way,[3,9] almost all the Hamiltonian methods of BCS-Eliashberg theory[14] can describe equally well the superconductive transitions of HTSC. Only the normal-state transport (not thermal) properties are affected by the marginal dimensionality of the layered structures because the scattering from residual (localized) defects obeys the extended times local equal local selection rule.[3,9]

IV. CALCULATION OF ELECTRON-PHONON INTERACTIONS

The close parallel between QPT and BCS-Eliashberg theory established by the selection rules described in Sec. III suggests that it should be possible to estimate the strength of electron-phonon interactions in QPT. We do this here using a simple model in which λ_p, the electron-phonon coupling strength, is approximated by an extension of weak-coupling BCS theory, where it is identified with $N(0)V$, where $N(0)$ is the density of states at $E=E_F$ and $V=\langle V_{\mathbf{k},\mathbf{k}+\mathbf{q}} \rangle$ is a suitably weighted average of the electron-phonon scattering matrix element. Our discussion necessarily contains several unknown parameters, and its aim is not the exact calculation of λ_p. Rather we seek to show that the usual limitations on λ_p set by band theory do not apply in the context of marginal dimensionality[1] and quantum percolation.[15]

Band theory normally establishes limitations on λ_p in two limits: the free-electron limit and the tight-binding limit. In the free-electron limit $\lambda_p=0$ and λ_p is small in nearly-free-electron metals (such as the alkalies[16]). In transition metals, where the d bands can be described by the tight-binding approximation,[17] $\lambda_p \lesssim 1$. This apparently leaves little room for $\lambda_p \sim 3-4$ which is needed[18] to explain $T_c \sim 90$ K in Y-Ba-Cu-O.

In QPT extended states are hybrids of ballistic states (similar to Bloch states) which are confined to domains in a single cuprate plane, with resonant interplanar defect states which pin E_F. Let us denote these states by ψ_b and ϕ_r, respectively, and represent an extended state ψ_e as a superposition of ballistic and resonant states

$$\psi_{en} = \sum_l a_{nl}\psi_b^l + \sum_m b_{nm}\phi_r^m \ . \tag{1}$$

Because the extended states ψ_{en} coexist[6] with localized states $\psi_{ln'}$ of the same energy, the phases χ_n^{ln} defined by

$$a_{nl}/b_{nm} = |c|\exp(i\chi_n^{lm}) \tag{2}$$

are physically significant; for the extended states χ_n^{lm} must correspond to traveling (not standing) waves.

In the band case we may write schematically for the electron-phonon interaction

$$\langle(\psi_b^l|\mathbf{u}\cdot\nabla W|\psi_b^{l'})\rangle = V_b \ , \tag{3}$$

where $\mathbf{u}$ is the phonon amplitude and W is the self-consistent screened ionic potentials. Different expressions are obtained for V_b depending on whether one represents ψ_b by pseudopotential[19] or tight-binding[17] wave functions, which do not concern us here.

In the percolative case the density of states near E_F has two components,

$$n(E)=n_b(E)+n_r(E) \tag{4}$$

associated with the Bloch-like ballistic intraplanar states and the resonant interplanar states. Although the total number of the latter is small, they are concentrated in a narrow range ($\lesssim 0.05$ eV) near E_F, compared to the bandwidth $B \sim 2$ eV for the relevant ballistic states. Thus we expect that $n_r(E_F) \sim n_b(E_F)$.

Does the increase in $n(E)$ due to the presence of $n_b(E)$ in (4) mean a large increase in $\lambda_p = n(E_F)V$? At first sight the answer to this question seems to be negative. This is because in

$$\langle(\psi_e^n|\mathbf{u}\cdot\nabla W|\psi_e^{n'})\rangle = V_e$$

there are three kinds of terms. We have the direct terms of ψ_b with itself, of ϕ_r with itself, and the cross terms. The direct ψ_b terms yield

$$\left\langle \left[\sum_{n,l} a_{nl}\psi_b^l \left| \mathbf{u}\cdot\nabla W \right| \sum_{n,l'} a_{nl'}\psi_b^{l'} \right] \right\rangle$$

$$= V_b n_b(E_F)[n_b(E_F)+n_r(E_p)]^{-1} . \quad (5)$$

The second factor on the right-hand side of (5) arises from the normalization of ψ_e^n. The direct terms of ϕ_r with itself should be small because the defect states have little overlap. The cross terms between ψ_b and ϕ_r can have either sign and so by symmetry (or the random-phase approximation) should average to zero. We are thus left only with (5), and when this is multiplied by $n_b(E_F)+n_r(E_F)$ to obtain λ_{pe}, we find that it is the same as λ_{pb}, namely $n_b(E_F)V_b$.

The essence of QPT is that it is a two-component Fermi-liquid model with both extended states ψ_e and localized states ψ_l, and the discussion just given concerned only ψ_e. When we consider ψ_l, the localized states, it seems that these may provide a source of broken symmetry, so that the cross terms involving ψ_e and ϕ_r do not cancel, but instead become phase coherent.[20] If this is the case, then we should first have some indication of broken symmetry, probably as a precursor effect due to strong electron-phonon coupling and lattice reconstruction.

Recently anomalies in ion channeling have been observed[21] that provide strong evidence for just such a precursor effect, which starts above $T_l \sim 120$ K in Y-Ba-Cu-O. Channeling experiments measure departures from planarity of the atomic sites, and the striking aspect of these data (measured with temperature decreasing) is that the disorder of the planarity of the oxygen atoms decreases rapidly starting near 120 K. Such a *reduction* in disorder cannot be caused by beam damage. It seems likely that it reflects the onset of some kind of local superconductive fluctuations which favor (and are favored by) oxygen planarity. This is consistent with the phenomenology of HTSC and layering. It has already been noted[22] that the buckling of the CuO_2 sheets decreases ($T_c = 90$ K) from Y-Ba-Cu-O to $Bi_2Sr_2CaCu_2O_8$ ($T_c = 85$ K) to Tl_2 $Ba_2CaCu_2O_8$ ($T_c = 110$ K), which suggests a positive correlation between decreased buckling and higher T_c.

Independent evidence for precursive local superconductive fluctuations is provided by electron-loss experiments.[23] The interpretation of these data is complicated by surface cleavage and electron-beam damage, but the loss edge near 45 meV which persists to $T \gtrsim 120$ K (well above $T_c \sim 90$ K) is still related to local superconductivity.[23]

If local superconductivity is present, then the cross-terms between ψ_b and ϕ_r need not average to zero, but instead may attain their maximum value in certain materials. (Near-attainment of an upper bound for T_c in ballistic metallic superconductors has been shown.[24]) It is easy to show from the golden rule that the overlap interaction energy S between ϕ_r and ψ_b is approximately given by $S^2 = B\Gamma$, where $B = 2$ eV is the ballistic bandwidth and $\Gamma = 0.05$ eV is my estimate of the resonant peak width. This gives $S = 0.3$ eV. If we now assume $n_r(E_F) = 2n_b(E_F)$, we have for the overlap contribution to λ a value $2S[n_b(E_F)+n_r(E_F)] = \lambda_{br} \sim \lambda_b$. This means that $\lambda_p = \lambda_b + \lambda_{br} \sim 2\lambda_b$.

The simple estimates we have just given for λ_p and λ_b are not true upper bounds. Even for the elemental metallic and binary intermetallic superconductors it is known[25] that experimentally $\lambda_b^{\max}$ is greater than or equal to 2. Presumably this is due to local-field corrections which are omitted from the simple estimates that give $\lambda_b \sim 1$. However, just as $\lambda_p \sim 2\lambda_b$, so we expect $\lambda_p^{\max} \sim 2\lambda_b^{\max} \sim 4$ when these corrections are taken into account. A value of $\lambda_p = 4$ is enough to explain $T_c \sim 100$ K with $\omega_D \sim 300$ cm^{-1} according to standard strong-coupling theory.[26] It also gives E_g/kT_c in the range 6–7, which seems to be the value most in agreement with a variety of optical, tunneling, and NMR relaxation data.[27]

V. CONCLUSIONS

We have seen that percolation, by itself, does not lead to enhancements of λ_p over the ballistic value λ_b found in intermetallic compounds. Instead local superconductive fluctuations must also be present to produce the broken symmetry that can generate phase coherence between the ballistic and resonant components of extended states. Such fluctuations are observable as a precursor effect through the temperature dependence of the oxygen planarity as measured in ion-channeling experiments.[21] Increased planarity corresponds to reduced intraplanar disorder, as already described in my earlier discussion of the existence of extended states in the context of marginal dimensionality.[1] It therefore seems that a consistent theory of HTSC, well supported by recent experiments, is emerging in the context of quantum percolation. Further evidence for precursive local superconductivity is planned to be discussed elsewhere.[9]

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