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Quantum percolation and lattice instabilities in high-T_c cuprate superconductors

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Lattice instabilities are the mechanisms that place upper bounds on T_c in theoretical models of superconductivity based on dynamical electron-phonon interactions. In percolative metals this upper bound can be much larger than in ballistic metals. For parameters appropriate to high-T_c cuprates, I show that the maximum value $\tilde{\lambda}_{\max}$ of the percolative electron-phonon coupling constant is about four times larger than the value $\lambda_{\max} \sim 1$ appropriate to ballistic metals. I also review values of λ in "anomalous" ballistic metals, such as Nb_3Ge homoepitaxial films, and show that these may be defect enhanced.

I. INTRODUCTION

In the standard Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity, electron pairing occurs because of a dynamical attraction between electrons caused by phonon exchange. The strength λ of this exchange is given by $N(E_F)V$, where $N(E_F)$ is the density of electron states at E_F, and V is an electron-phonon interaction energy. When this attractive interaction was first proposed,[1,2] it was immediately noticed[3] that with increasing λ the lattice would become unstable,[4,5] qualitatively for $\lambda > \lambda_{\max} \sim 1$. With superconductive tunneling it became possible to evaluate λ for materials which would form good tunnel junctions, and to check these values with specific-heat data. It was found that on the whole this qualitative argument was correct. There are some chemical variations in $\lambda_{\max}$ for simple *s-p* metals, transition metals, and transition-metal–metalloid alloys, but most of these are well understood. The general background for this subject is reviewed in my book.[6] One minor problem, that of Nb_3Ge, was left unresolved by the extensive discussions of lattice instabilities in the context of tunneling experiments, neutron measurements of lattice vibrations spectra, and extensive electronic band-structure calculations that were carried out[7,8] in the 1970's. For the reader's convenience I review this material, as well as the formal literature as it developed up to 1980, in Sec. II. There I also suggest a simple explanation for the high T_c's observed occasionally in Nb_3Ge.

The main aim of this paper is to begin the discussion of the analogous condition for $\lambda_{\max}$ in high-T_c cuprates, where there is a growing experimental evidence for my model of conductive electronic states which are not ballistic but are percolative on an atomic scale. The general motivation for this model is discussed in my book,[6] and some of the more recent data are reviewed in this context in Sec. IV. This model is entirely new and probably will not be familiar to those who have not read my book or the background papers on localization which I have written in the 1980's. The discussion of the conceptual basis of my model in Sec. III is necessarily condensed, and the reader who wishes to understand these ideas in detail is well advised to consult the earlier references.

The general problem which confronts any percolative model, classical or quantum, is that the percolative states cannot be simply identified. For ballistic conduction by plane-wave-like states indexed by crystal momentum **k**, and with a group velocity $\mathbf{v} = \nabla_k E(\mathbf{k})$ obtainable from one-electron band theory and renormalizable (in principle, at least) by nearly free-electron many-particle techniques, electron-phonon interactions can be (and have, especially for simple metals like Al) calculated quite accurately.[6] For percolative conduction, this is no longer the case. Thus, although experimental evidence is indeed accumulating which supports the percolative model (Sec. IV), it is still not clear how strong the electron-phonon interaction at defects can be, that is, what $\lambda_{\max}$ is in the percolative case.

The view taken in Sec. III is that we will explore several models for $\lambda_{\max}$ in order to see how it may differ in the percolative from the ballistic case. By so doing, we will be able to avoid the apparently intractable problems associated with identifying the specific defects and their configurations and then finding the electronic states which actually percolate, and how these are scattered by lattice vibrations. Instead we simply assume that the values of $\lambda_{\max}$ calculated in a percolative model will correspond fairly well to those achieved in the highest-T_c cuprates. This is a reasonable assumption, which has already been verified in the metallic cases, even, as we shall see, in the so-called "unsolved" case of Nb_3Ge. It is therefore a reasonable goal for theory in the cuprate case.

II. LATTICE INSTABILITIES IN BALLISTIC METALS

By a ballistic metal I mean a material with metallic conductivity $(d\rho/dT > 0)$ where the current-carrying electrons (or holes) can be identified with quasiparticles of crystal momentum **k** and (if necessary) a Bloch band index n. The connection between electron-phonon coupling strength λ, electron-ion screening, and lattice instabilities has been discussed in many papers. In a jellium or continuum model one can easily estimate[3–5] that $\lambda_{\max} \sim 1$, because for larger values of λ the electrons "overscreen" the ions and the lattice becomes unstable.

Real solids, of course, have definite crystal structures with atoms with complex atomic potentials at the atomic sites. In this case, in addition to the continuum screening terms, one has umklapp screening involving reciprocal-lattice vectors $\mathbf{G} \neq 0$ and the local-field corrections associated with this screening should be considered, as well as the nonlocal atomic-pseudopotential form factors. For polyatomic unit cells one must treat optic modes as well as acoustic modes. These corrections greatly complicate the lattice-dynamical problem, and as a result the electron-phonon interaction strength λ for scattering on the Fermi surface is no longer simply related to lattice instabilities, especially if these involve optic modes, which is usually the case when there is more than one atom per unit cell. Even in the simple case of acoustic modes, one can easily use standard self-consistent-field screening techniques[9] to show[10,11] that, in principle, λ might be very large before the lattice becomes dynamically unstable. These techniques, however, consider only instabilities due to second-order phase transitions. Indeed once $\lambda > \lambda_{\max}$ in polyatomic crystals, many first-order phase transitions can occur which may include phase separation.

By now more than 70 superconductors are known[6] with $T_c > 10$ K, and for many of these we have estimates of T_c from tunneling and specific-heat data,[12] as well as band calculations of λ based on various approximate methods.[13,14] This means that we can test the notion that a $\lambda_{\max}$ exists simply by surveying known values and grouping the materials chemically and structurally. The advantage of this approach is that it includes not only second-order displacive instabilities (which are calculable, in principle, by perturbative methods, even though it seems that this has seldom been done in practice), but also first-order instabilities (such as phase separation) as well.

Suppose we consider a close-packed metal, like Al, which has a nearly free-electron band structure and a small atomic core. In this case theory can calculate T_c and λ accurately because both are small (1.2 K and 0.43, respectively). We can[13] increase T_c by increasing the ion-core charge Z and decreasing the valence-electron density n, as in Pb, the result being $T_c = 7.2$ K and $\lambda = 1.5$ (expt.) or 1.3 (theory, omitting nonlocal corrections). We notice that already $\lambda > 1$, presumably because a close-packed structure is inherently more stable than jellium. (This is an important observation, because the cuprate structures are based on the open perovskite structure and contain many vacancies as well. Thus, if these materials were ballistic metals they would probably already be mechanically unstable at $\lambda = 1$ or less.) Continuing along this line we form $Pb_{1-x}Bi_x$ alloys,[14] increasing T_c and λ to 9.0 K and 2.1 at $x = 0.35$. For larger x a nonclassical transition from a dense close-packed structure to the open, nearly simple-cubic structure of Bi with a miscibility gap of $\Delta x = 0.65$ and phase separation occcurs.[14]

We see from this example, which is a case where the exception proves the rule, that $\lambda_{\max} = 1$ is a good criterion for most simple metals which do not have close-packed structures. For transition metals the largest value is $Nb_{0.75}Zr_{0.25}$ ($T_c = 10.8$, $\lambda = 1.3$), which is similar to Pb. To go to higher T_c, one must use binary transition metal-metalloid compounds.

Before discussing binaries (and later ternaries, quaternaries, and quinaries for the cuprates), it is important that we pause to orient ourselves. In cubic monatomic elemental materials and pseudoelemental alloys (such as Tl-Pb-Bi) the crystal structure contains only one coordinate, the lattice constant. In polyatomic multinary compounds there can be many internal coordinates in the unit cell, and associated with each of these, lattice instabilities can occur. [Perhaps the simplest (but most often overlooked) of these is the antisite defect.] Therefore, we must be careful in generalizing results on λ from elemental monatomic systems to polyatomic multinary ones. (It was just this generalization that was made by McMillan in his empirical fits[15] to tunneling values of λ which led him to conclude that $T_c(\lambda)$ saturates with increasing λ, whereas it is now known[16] that $T_c(\lambda)$ for large λ increases like $\lambda^{1/2}$.) Some polyatomic multinary systems behave like monatomic elemental ones, while others do not.

A useful global classification of superconductive crystal structures for our purposes has been made[17] using Villars quantum structure diagrams based on quantum-mechanical definitions of size and electronegativity. The plot for high-T_c superconductors is shown for the reader's convenience in Fig. 1. On these plots a suitably weighted size differences $\overline{\Delta R}$ is the single most important variable, although the other variables, such as average valence number $\overline{N}_v$ and electronegativity differences $\overline{\Delta X}$ are important as well. On the $\overline{\Delta R}, \overline{\Delta X}$ plot the pseudoelemental systems, such as Tl-Pb-Bi, obviously fall near the origin, in what is called[17] island A. This small island contains mainly A15 compounds, which thus might be described, like the Pb-Bi system, as having high T_c's because they are vicinal to a covalent-metallic instability.[18] However, this description need not apply[6,17] to island B (NbN family) or to island C (Chevrel compounds like $PbMo_6S_8$ or high-T_c cuprates).

At this point it is convenient for us to examine, and in my opinion resolve, a long-standing puzzle, which is the anomalous behavior of T_c in Nb_3Ge. It turns out that the resolution of this puzzle forms a useful introduction to the quantum percolation model which is the main subject of this paper.

The high-T_c materials with the A-15 structure that are easily prepared are V_3Si ($T_c = 17$ K) and Nb_3Sn ($T_c = 18$ K, $\lambda = 1.7$). Stoichiometric Nb_3Ge has not been grown as large single crystals, but for purposes of neutron scattering a 0.1-cm^3 sample of $Nb_{3.2}Ge_{0.8}$ ($T_c = 6$ K) was prepared,[19] with 20% of the Ge sites occupied by Nb, which reduces T_c from the value of 17–20 K typical of most polycrystalline and thin-film samples.[20]

The lattice constant a of Nb_3Ge can be expanded in two ways. Bombardment by α particles[20] increases a by

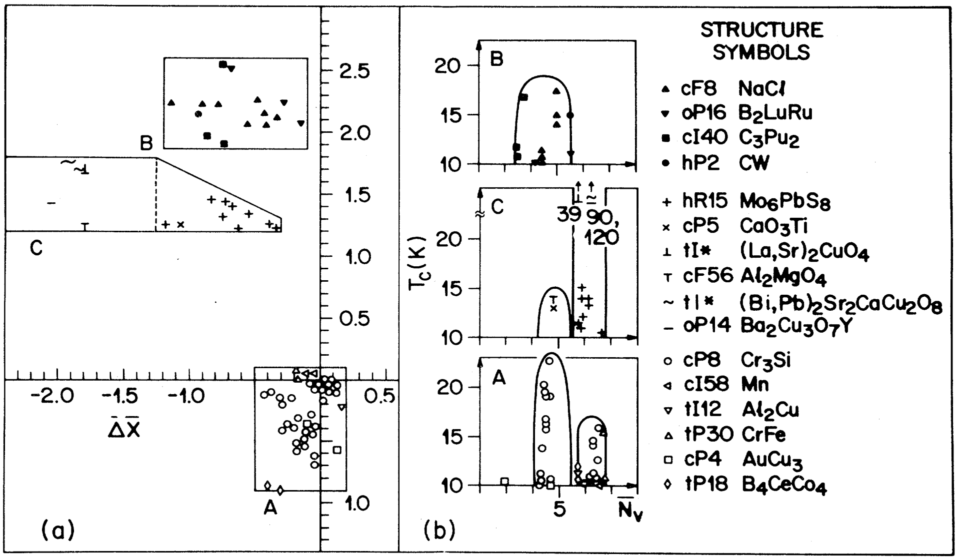

FIG. 1. Diagrammatic separation of high-T_c superconductors according to type of lattice instability (see Ref. 17). The diagram is essentially complete: it includes ternaries as well as binaries (note the crystal structure types listed on the right), and the separation using chemical variables defined quantum mechanically (rather than empirically) is essentially perfect.

1% and decreases T_c from 20 to 4 K. On the other hand, by using a suitable $A15$ substrate a 1 or 2% larger lattice constant can be imposed on a homoepitaxial thin film. This has the effect of expanding the range of thermodynamic stability[21] of $Nb_{1-x}Ge_x$ in the A-15 structure from $x=0.19$ to 0.26. Although the exact composition of the film is not known, this appears to produce a maximum in $T_c(x)$ near $x=0.25$. This enhanced stability and sample volume filling factor of the A-15 phase of Nb_3Ge with an expanded lattice constant was confirmed on a different substrate.[22]

The fact that T_c can be enhanced by nudging a favorable phase closer to a lattice instability is, of course, fully in line with our general expectations. The theoretical problem arises when we compare the electronic structure of Nb_3Ge with that of Nb_3Sn calculated by the same method.[23] The calculated value of $\lambda=N(E_F)V$ in Nb_3Ge appears to be lower than in Nb_3Sn, because V should be changed little and near E_F the upper peak in $N(E)$ seems to be ~ 1.5 (unit cell eV)$^{-1}$ in Nb_3Ge compared to 1.9 (unit cell eV)$^{-1}$ in Nb_3Sn, as shown in Fig. 2, reproduced from Ref. 23 for the reader's convenience. Are these discrepancies explicable?

First we note that $N(E_F)$ estimated from specific-heat data for Nb_3Ge should be low, because of heterophase problems which almost surely exist even when the minority phases are not detected by diffraction on homoepitaxial films. The theoretical estimate of $N(E_F)$ could arise from band-structure errors on a scale of 0.05 eV, which is an accuracy well beyond the present state of the art on an absolute basis. However, one may ask why these do not spoil the Nb_3Sn calculation. This could be because of a fortuitious choice in atomic sphere sizes for Nb and Sn (which improves convergence in Nb_3Sn but not for Nb and Ge). Since there is no general rule here, the choice of sphere sizes is arbitrary, and a sharpening of $N(E)$ near $E=E_F$ in Nb_3Ge in a more completely converged band calculation is the easist way to resolve this puzzle.

Another way is the presence of 1–2 % Ge atoms in Nb sites in the expanded films grown epitaxially.[21,22] Because the Ge atoms do not have d states near E_F their presence on Nb chains, which have been weakened by a lattice constant expansion, could enhance the upper peak near E_F in Nb_3Ge to make it even stronger than in

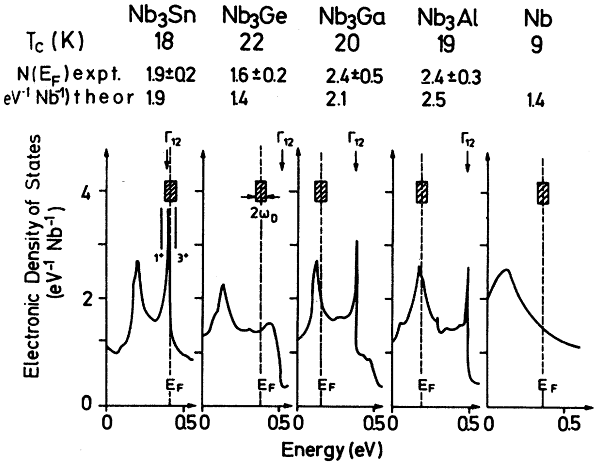

FIG. 2. Summary of density of electronic states $N(E_F)$ for various high-T_c superconductors (idealized crystal structures) calculated theoretically, and also measured experimentally by specific heats, reproduced from Ref. 23 for the reader's convenience.

Nb_3Sn, and bring it closer to E_F. Such enhancement generally occurs for a short-range potential whose strength is just below the critical value needed to break off a discrete state from a band edge.[24,25]

I have discussed the case of Nb_3Ge in some detail because one often hears casual comments to the effect that the Nb_3Sn-Nb_3Ge discrepancy illustrated in Fig. 2 is "unexplained," and that this somehow is indicative of an overall inadequacy of the BCS theory. These comments are often made by individuals with little or no detailed understanding of the materials problems associated with homoepitaxial films or the computational problems associated with one-electron self-consistent-field calculations for materials with many atoms per unit cell. From the discussion we see that the "discrepancy" in Fig. 2 can be explained in at least two ways, because both the experiments and the theory are technically very difficult. These same technical difficulties are present to a larger degree in the high-T_c cuprate superconductors. The same problems arise: heterophases, defect enhancement of $N(E_F)$, and convergence limitations of the band calculations. However, if we are aware of these technical difficulties, we will be less likely to regard them as fundamental failures in principle, and therefore may be less tempted to use them as a justification for revisionist exotic theories.[6]

III. MAXIMUM ELECTRON-PHONON COUPLING STRENGTH $\tilde{\lambda}_{\max}$ IN PERCOLATIVE METALS

The salient characteristic which suggests percolation in high-T_c cuprate alloys is that very small changes in composition or structure (sometimes too small to be detected by diffraction) can change the material from semiconductive ($d\rho/dT < 0$ for T above T_c) to metallic ($d\rho/dT > 0$).[6] In the best samples ρ becomes a linear function of T (sometimes over several decades), and this behavior is also explained (without adjustable parameters or atomic form factors) by the quantum percolation model.[26]

In most materials percolative conductivity arises as the result of the presence of at least two phases, with a minority metallic phase dendritically embedded in a majority semiconductive or insulating phase. The cuprates are special because they contain, in effect, semiconductive and metallic phases on an atomic scale. Moreover, according to the model, even the "metallic" cuprate planes are actually not metallic in isolation, but they become so only when rendered effectively three dimensional through connection by interplanar metallic defects embedded in intervening semiconductive planes.[27]

The picture of electrical pathways in this model is that they consist of domains of CuO_2 planes (usually) or CuO chain segments (occasionally) alternating with interplanar defects with a density $f = n/N$, where n is the number of defects per planar domain or chain segment of N atoms. Electron-phonon coupling occurs with a strength λ_1 in the planes, and a strength λ_2 at the defects.

We now ask what the mechanical consequences of these two kinds of couplings should be. First note that as the defect density $f \to 0$, the planes become semiconductive well before they become antiferromagnetic.[6] The simplest way to explain this, in accord with earlier reasoning about the separability of localized and extended states,[28] is to divide the planes and chains into domains, with one domain per planar defect and one extended state per excess of axial defects over planar defects. If we assume that the electron-phonon coupling for localized states λ_l is small ($\lambda_l < 1$) and similar to Al ($\lambda_l \sim 0.5$), then the degrees of freedom associated with these localized states can act as a "cage" to restrain the instabilities associated with the strong interactions of the extended states. This suggests that we replace the ballistic condition

$$\lambda < \lambda_{\max} \sim 1 \tag{1}$$

by a percolative condition, normalized per extended state. This would be

$$f\lambda_1 + (1-f)\lambda_l \lesssim 1 \tag{2}$$

or, with $\lambda_l = \frac{1}{2}$ and $f = 0.1$ (as in $La_{1.8}Sr_{0.2}CuO_4$),

$$\lambda_1 \lesssim 5.5 \ , \tag{3}$$

which means that in the percolative case

$$\tilde{\lambda}_{\max} = 5.5\lambda_{\max} \ , \tag{4}$$

which is a sufficiently large enhancement of the coupling strength to explain the high-T_c's in the cuprates.

Conditions (3) and (4) apply to the planar instabilities. The "cage" that stabilizes the defects, which are embeded in the semiconductive layers, is probably more rigid than that which stabilizes the CuO_2 planes. This suggests that the electron-phonon coupling λ_2 at the defects may be much larger than the coupling λ_1 in the planes. On an octahedral nearest-neighbor basis, $\frac{2}{3}$ of each cage is coplanar, while $\frac{1}{3}$ is in the nearest-neighbor planes, so that λ_2 might be as much as $2\lambda_1$. Experimental evidence on these points is discussed in the next section.

Although (4) is a satisfying result, there are other possibilities. The most important of these is that as f increases the samples may become severely inhomogeneous (incipient phase separation). In general, no analytic technique exists for discussing phase separation, but intuitively one would like to think that this would arise as a result of defect-defect coupling mediated by CuO_2 planes or CuO chains. Such a coupling would itself involve mechanical percolation from defect to defect, which would depend on the magnitudes of λ_1 and λ_2. A simple expression which does this is

$$f\lambda_1\lambda_2 \lesssim 1 \ , \tag{5}$$

which might be regarded as an alternative to (4). To maximize T_c we would like to have $\lambda_1 = \lambda_2 = \text{const}$ in which case (5) becomes

$$\tilde{\lambda}_{\max} = f^{-1/2} \sim 3.5 \tag{6}$$

for $f = 0.1$. For heuristic purposes, there is little difference between (4) and (6), because they both show that enhancement of $T_c(\lambda)$ from 20 K (1.5) to 125 K (4) is possible because of the cage effect.

Some comments should be made on the justification for Eqs. (2) and (5). Neither is based on a normal-mode analysis, but this problem is less serious than it may appear. First, we know from studies of percolation of mechanical instabilities in noncrystalline networks[29] that a mean-field approximation, such as (2), is much more accurate for mechanical instability percolation than it is for electrical percolation. Second, with such large unit cells the crystalline periodicity itself becomes of secondary importance and the strength of the coupling at which the structure becomes unstable more closely approaches that of a continuum model. [The largest uncertainty in using Eq. (2) actually lies in the estimate of the strength λ_l of the cage.] Finally, the defect-medium-defect interaction described by (5) is the simplest model which could lead to defect "precipitation" and phase separation. Like the mean-field model (2), it is not likely to be seriously in error.

IV. COMPARISON WITH EXPERIMENT

The equations (2) and (4) which define $\lambda_{\max}(f)$ determine only a bound on λ and T_c which may not be attained. Broadly speaking, T_c is observed to increase with dopant concentration x (for example, in $La_{2-x}Sr_xCuO_4$) in several high-T_c cuprates. However, above a certain critical concentration some kind of lattice instability (oxygen vacancies, phase separation, or rapid increase in heterophase character) limits T_c. It is this second effect which our discussion is meant to describe. Both effects are illustrated in Fig. 3.

It is important to realize that there is still some uncertainty about f in (2) and (4), and that it may be close to, but is not necessarily equal to, x in p-type $La_{2-x}Sr_xCuO_4$. The same uncertainty applies to n-type alloys such as[30,31] $Nd_2CuO_{4-x}E_x$ and $Nd_{2-x}Ce_xCuO_{4-y}$. In these materials $T_c \sim 30$ K. In materials such as $YBa_2Cu_3O_7$ and the bismates and thallates,[6] $T_c \sim 100$ K and it seems likely that f is larger, but we have no firm evidence on this point. Attempts to determine f directly by Hall-effect measurements near T_c may not yield reliable values because of macroscopic percolation effects which are sensitive to temperature-dependent resistivity anisotropy ratios.[26]

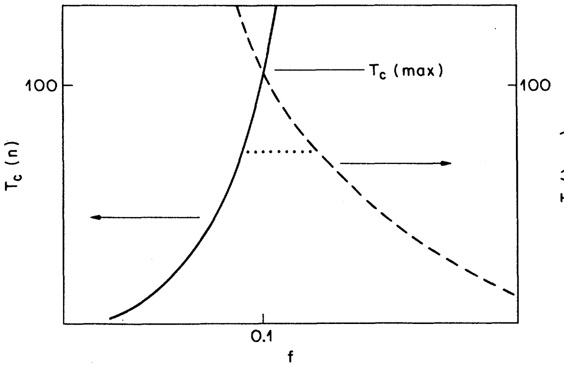

FIG. 3. Sketch of the two competing factors which determine the maximum value of T_c: one is the density n of percolating states, while the other is lattice instabilities. Both are related to f, the fraction of interlayer defects discussed in the text.

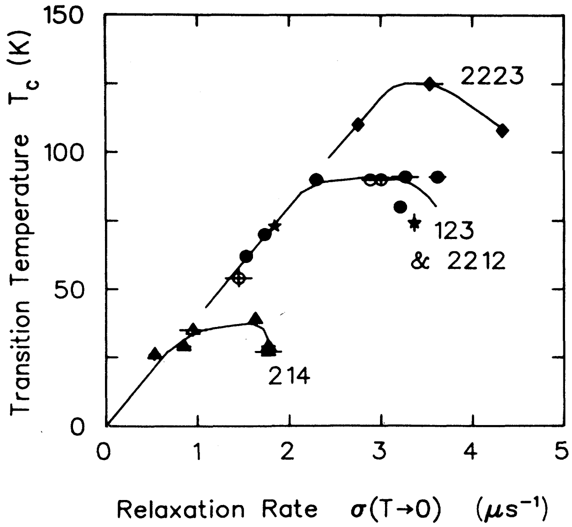

FIG. 4. Correlations between T_c and σ (muon-spin-relaxation rate) for cuprate superconductors, reproduced here for the reader's convenience from Ref. 32.

Perhaps the best way to examine the relation shown in Fig. 3 is by measuring[32] the muon-spin-relaxation rate σ. In homogeneous type-II superconductors σ is directly proportional to the carrier density n_s (or f in Fig. 3). In percolative superconductors where only an unknown fraction of the sample volume becomes superconductive, and the material is electronically inhomogeneous, this simple relation no longer holds.[33] What this means in practice is that qualitative trends can probably be inferred correctly, but that the quantitative functional form which is measured is probably distorted by hidden composition-dependent filling factors from the general trends shown in Fig. 3. This is evident in Fig. 4, which is reproduced from Ref. 32. The breaks in the slope of $T_c(\sigma)$ are the results of changes in the filling factors and associated spinodal electronic phase separation, which initially limits T_c and for large σ eventually decreases it as the fraction of superconductive electronic phase decreases and percolation is suppressed. The spinodal region is indicated in Fig. 3 by the dotted line.

With these uncertainties in mind we can see that the present discussion merely provides a useful supplement to experimental data which indicate that for small doping T_c increases with x. We can estimate how far this increase continues before it is stopped by a lattice instability by converting our estimates of $\lambda_{\max}$ into estimates of T_c by using an Einstein model for the phonon spectrum and various solutions to the Eliashberg equations which have been derived both analytically[33] and numerically.[34] For the phonon energy we choose $\hbar\omega_D = 300$ cm^{-1} [experimental values of the CuO_2 planar oxygen A_{1g} (z polarized) bending mode range from 340 cm^{-1} in $YBa_2Cu_3O_7$ to 280 cm^{-1} in the bismates[6]]. Using the equations[33]

$$T_c = 0.25\omega_p (e^{(2/\tilde{\lambda})} - 1)^{-1/2} , \tag{7}$$

$$\tilde{\lambda} = \lambda (1 + 2.6\mu)^{-1} , \tag{8}$$

a largish Coulomb repulsion parameter $\mu = 0.2$, and $\lambda = 4.5$, we have $\tilde{\lambda} = 3.0$ and $T_c \sim 110$ K, which is satisfactory. Without the cage effect the closest analogy with the cuprates is either the A-15 (Nb_3Sn) or B-1 (NbN) families where $T_c \lesssim 20$ K.

In conclusion, our analysis shows that the quantum percolation model with very strong ($\lambda \sim 5$) electron-phonon coupling in CuO_2 planes is capable of explaining the observed T_c's in the cuprates. This strong electron-phonon coupling should lead to buckling of the CuO_2 planes. This has indeed been observed,[36] and when the cage is slightly stronger, so that the buckling is reduced, as in the $YBa_2Cu_3O_7$, bismate, thallate series, T_c is increased,[36,37] as one would expect with a larger $\tilde{\lambda}_{max}$. This reasoning strengthens previous general arguments[6] that exotic models are unnecessary for the explanation of high-T_c superconductivity.

ACKNOWLEDGMENTS

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