Theory of the "universal" degradation of T_c in high-temperature superconductors

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The superconducting T_c decreases similarly with disorder (as measured by resistivity ρ) for all systems with high T_c. We propose that this is a localization effect. For strong disorder, electron diffusion is slow and strongly scale dependent. This increases the effective Coulomb repulsion. Model calculations with realistic parameters describe well the experimental T_c-vs-ρ curves. Explanations are suggested for the relatively large critical conductance found.

It is well known that high-transition-temperature superconductors are unusually sensitive to disorder.[1] For example, the transition temperature T_c of Nb_3Ge, an $A15$ structure material, drops from 22 to about 5 K as its resistivity ρ increases from 40 to 130 $\mu\Omega$ cm. It has been increasingly realized that resistivity is the single "universal" measure of damage or disorder relevant for T_c. Irrespective of how disorder is caused, e.g., by particle bombardment or alloying, there is one T_c-vs-ρ curve. Studies on $A15$'s such as V_3Si, Nb_3Sn, and Nb_3Ge as well as on the high-T_c cluster compounds $LuRh_4B_4$ and $ErRh_4B_4$ show similar dependence of T_c on ρ except for scale changes.

The conventional view of this striking effect is that the high T_c is due to a sharp peak in the density of states $N(\epsilon)$ near the Fermi energy ϵ_F. This is progressively smeared with increasing disorder, thus reducing $N(\epsilon_F)$ and thence T_c.[2] This kind of one-electron description is unrealistic, since electron-phonon coupling effects are strong in these systems, as is clear from giant thermal resistivity, large phonon anharmonicity, and small Debye-Waller factor, etc. The peak in the single-particle density of states is reduced in height as well as being rounded due to these electron-lattice coupling effects. Further, since the Cooper pair attraction is over the Debye energy range, i.e., over $|\epsilon-\epsilon_F| \lesssim \hbar\omega_D$, this averaging also reduces the effect of a peak in the density of states. Another argument against a one-body explanation is the fact that most of the T_c decrease occurs for $\rho \gtrsim 40$ $\mu\Omega$ cm where the mean free path is at a most few lattice spacings. It is not clear that more disorder will further smear the one-electron density of states. More directly, tunneling experiments on Nb_3Al (Ref. 3) show that the density of states remains nearly constant up to $\rho \sim 110$ $\mu\Omega$ cm while T_c falls by a factor of 4. Also, not all high-T_c materials have a large electronic specific heat coefficient γ.

We propose here that Anderson localization in extremely disordered systems provides a natural explanation for a universal degradation of T_c. Based on the scaling theory results of Abrahams *et al.*,[4] we show that the Coulomb pseudopotential μ^*, i.e., the effective Coulomb repulsion between Cooper pair electrons, increases with disorder. The screened interaction μ is of short range in space and is nearly instantaneous on a time scale of order ω_p^{-1} where ω_p is the plasma frequency. For strong disorder, electron density fluctuations diffuse very slowly; over a considerable length scale the diffusion constant $D(L)$ *decreases* with increasing scale size L as L^{-1} (Ref. 4). Thus density fluctuations interact more strongly, and with characteristic retardation so that $\mu(\omega)$ increases with decreasing frequency ω. We calculate this as well as the consequent increase in μ^* below. The retardation effect is important since a mere increase in the instantaneous μ changes

$$\mu^* = \mu/[1+\mu \ln(\epsilon_F/\omega_D)]$$

only slightly. When the disorder is nearly critical (i.e., the Fermi energy is close to the mobility edge), the macroscopic resistivity ρ is (much) larger than the microscopic short-length scale resistivity, which is close to the critical value ρ_c. In this regime, we show that the change in μ^* is essentially a function of ρ/ρ_c. We argue that of the quantities which determine T_c, μ^* is most sensitive to strong disorder so that in this regime ($k_F l \sim 1$, where k_F is the Fermi wave vector and l the mean free path), T_c is a "universal" decreasing function of ρ/ρ_c. A frequency-dependent enhancement of the Coulomb vertex for $k_F l \gg 1$ where electron diffusion is fast and classical, i.e., scale independent, was first pointed out by Altshuler and Aronov.[5] The regime considered here is qualitatively different.

We briefly mention the weak-disorder ($k_F l \gg 1$) regime. It is known experimentally[6] and theoretically[7] that weak disorder has little effect on T_c. Gorkov[8] has shown that the pair density of states is unchanged to order $(k_F l)^{-1}$. As for the electron-phonon interaction λ, it is a general consequence of perfect screening and of the fact that random scatterers move with the lattice that the electron—long-wavelength-phonon vertex is unchanged by disorder.[9,10] In contrast, the Coulomb vertex is characteristically enhanced by disorder.[5] The effect of this dif-

fusion and localization on T_c has been considered perturbatively in this regime by Maekawa and Fukuyama[11] in two dimensions. The effect in three dimensions has not yet been calculated, but is expected to be of order $(k_F l)^{-2}$.

We now calculate the effective Coulomb interaction in the strong-disorder regime. The Coulomb interaction kernel $K^c(q,\omega)$ at wave vector q and frequency ω is given by

$$K^c(q,\omega)=V_{\rm scr}(q,\omega)P(q,\omega)\ , \tag{1}$$

where $V_{\rm scr}(q,\omega)$ is the dynamically screened Coulomb potential, and $P(q,\omega)$ is the probability of a density fluctuation n propagating with frequency ω, i.e., it is the Fourier transform of the density-density correlation function. Relation (1) is most simply obtained in an exact eigenstate representation,[12] and the factor $P(q,\omega)$ describes the diffusional correlation between electron density fluctuations interacting through the Coulomb potential. $V_{\rm scr}(r,t)$ is short range in space and nearly instantaneous, i.e., $V_{\rm scr}(q,\omega)N(\epsilon_F)=\mu$ so that the local Coulomb kernel

$$\sum_q K^c(q,\omega)N(\epsilon_F)=K^c(\omega)$$
$$=\frac{\mu}{\bar{n}^2 N(\epsilon_F)}\int dt\langle n(0,t)n(0,0)\rangle e^{i\omega t}\ .$$

Now quite generally, the density correlation function has a free-particle form for $t<\tau$ (collision time) and a diffusive form for $t>\tau$, so that

$$\langle n(r,t)n(0,0)\rangle=[\bar{n}^2/Q(t)]\exp(-r^2/4Dt)$$

for $t>\tau$. Here the diffusion constant D is, in general, a function of r and $Q(t)$ is determined by normalization of probability, i.e., by the condition

$$\int\langle n(\vec{r},t)n(0,0)\rangle d\vec{r}=\bar{n}^2\ .$$

The kernel is therefore

$$K^c(\omega)=\mu\left[1+\frac{2}{N(\epsilon_F)}\int_\tau^\infty\left[\frac{\cos\omega t}{Q(t)}\right]dt\right]\ , \tag{2}$$

where the first term on the right is the free-particle contribution. We now show that $Q(t)$ (the probability of finding that an electron has not diffused away in a time t) increases in a characteristic way with resistivity, due to localization effects.

In the scaling theory of localization, the dependence of conductance g on scale size L is described by a universal function $\beta(g)=d\ln g/d\ln L$. For large g, $\beta(g)$ can be calculated perturbatively[4] and the leading term is $\beta(g)=1-g_c/g$ for three dimensions, where $g_c=e^2/\hbar\pi^2$ for a free-electron gas. It has been shown that there are no new terms to order $1/g^3$.[13] Numerical simulation[14] also gives a slope of 1.2 ± 0.3 for $\beta(g)$ near its zero at $g_c\simeq e^2/2\hbar\pi^2$. We therefore assume this form for $\beta(g)$ to be reasonably accurate near g_c also. One has then $\sigma(L)=\sigma+g_c/L$, where

$$\sigma=\sigma(\infty)=\rho^{-1}=\sigma_0[1-3(k_F l)^{-2}]$$

as estimated by perturbation theory, and it goes to zero for critical disorder ($k_F l=\sqrt{3}$ perturbatively where l is the microscopic mean free path) so that $\rho\to\infty$ there. Up to a length scale $L_s=g_c/\sigma=g_c\rho$, the conductivity and the diffusion coefficient vary roughly as $1/L$. If the disorder is sufficiently strong so that $L_s\gg l$, this region is sizable, and approximately, $D(L)=D_0(l/L)$ for $l<L<L_s$ and $D(L)=D_0(l/L_s)$ for $L>L_s$, where $D_0=v_F l/3$ is the microscopic diffusion constant. This crude approximation brings out the differences in the normalization constant in the two regimes. One has $Q(t)=(16\pi/3)D_0\,|\,t\,|\,l$ for the nonclassical diffusion regime $\tau<t<(L_s/l)^3\tau=\alpha\tau$, and $Q(t)=(4\pi D_0\,|\,t\,|\,l/L_s)^{3/2}$ for longer times $t>(L_s/l)^3\tau$, where the diffusion is classical. In the nonclassical regime, $Q(t)^{-1}$ decreases only linearly with time because of the critical slowing down of electron diffusion compared to the normal $t^{-3/2}$ decrease. This increases the Coulomb repulsion, i.e., from Eq. (2) the kernel

$$K^c(\omega)=\mu[1+(9\pi/4)(k_F l)^{-2}\ln(\omega\tau)] \tag{3}$$

for $\alpha^{-1}<\omega\tau<1$, joining smoothly to obvious frequency-independent values for $\omega\tau>1$ and $\omega\tau<\alpha^{-1}$.

In order to calculate the change in T_c, the linearized gap equation must be solved including in addition to the kernel $K^c(\omega)$ the effect of strong disorder on the phonon-induced attraction λ, and on the pair propagator. The electron-phonon vertex relevant for the former involves low frequencies $\omega<\omega_D$ or long time scales. In this range the diffusion is expected to be classical if $\omega_D<(\alpha\tau)^{-1}$. When diffusion is classical, the electron-phonon vertex is not enhanced[9] for phonon wave vectors $q<l^{-1}$ due to the complete screening requirement mentioned earlier. Since in the present case $ql<1$ covers a considerable range in q space, we assume that there is no increase in the electron-phonon vertex. We also ignore model-dependent corrections to the BCS attraction from the region $ql>1$.[8]

The pair propagator is affected by disorder, especially in the presence of Coulomb and electron-phonon interactions. For example, the single-particle density of states has a dip[10] due to Coulomb interaction which becomes more pronounced near critical disorder.[15] There is also a reduction due to the Hartree diagram involving electron-phonon coupling.[16] Some of these effects have been considered perturbatively[11] in two dimensions; their effect is to reduce T_c as a function of resistivity. There is no reliable way, however, of calculating the pair propagator or the density of states near critical disorder. We assume that they do not change much, on the basis of experimental results which indicate[3] only small changes in the density of states for large disorder.

The main effect of proximity to localization is thus an increase in the Coulomb pseudopotential μ^*. This increase can be calculated by solving the linearized gap equation in the standard two square-well approximation, i.e., $\Delta(\omega)=\Delta_1$ for $|\omega|<\omega_D$ and $\Delta(\omega)=\Delta_2$ for $\tau^{-1}>\omega>\omega_D$. We find

$$\mu^*=\mu'/[1+\mu'\ln(\epsilon_F/\omega_D)-(\mu'-\mu)\ln(\sqrt{\alpha}\epsilon_F\tau)]\ , \tag{4}$$

where

$$\mu'=\mu[1+(9\pi/4k_F^2 l^2)\ln\alpha]$$

and

$$\alpha=(g_c\rho/l)^3=(\rho/\rho_c)^3\ .$$

To calculate T_c, we use McMillan's formula,

$$T_c=\frac{\omega_D}{1.45}\exp\left[-\frac{1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right]\ ,$$

where μ^* as a function of the resistivity is given in Eq. (3) and λ is the electron-phonon coupling. Because of the approximations made, μ^* has its classical value

$$\mu_0^*=\mu/[1+\mu\ln(\epsilon_F/\omega_D)]$$

up to $\rho=\rho_c$ beyond which localization effects increase it. Actually, μ^* increases continuously from μ_0^* as ρ increases.

We compare our results with experiment assuming that the systems considered are in the strong-disorder regime where the anomalous diffusion effects will be important. We assumed that λ and μ do not change with disorder. The values of λ and ω_D are taken from experiment where possible. Otherwise, numbers consistent with T_c and with normal-state properties are used. We take $k_F l=1$, $\mu=0.3$ for all the three systems, Nb_3Ge, V_3Si, and $LuRh_4B_4$, considered. Figure 1(a) shows the experimental T_c-vs-ρ curves (Ref. 1, Rowell and Dynes) and the results of Eq. (3). The constants used are shown. The only free parameter is ρ_c, which is in the range 7—31 $\mu\Omega$ cm, and is adjusted to give the best fit for large ρ. We do not expect a good fit for small $\rho\sim\rho_c$ because of the approximations used. We see that the data are well described over a factor-of-5 range in T_c. Equally good fits are obtained with ρ_c in the range 13—57 $\mu\Omega$ cm, and with $\ln(\epsilon_F/\omega_D)=5$ [Fig. 1(b)]. The values of ρ_c obtained correspond to a critical conductance $g_c=(l/\rho_c)\sim 10^{-3}\ \Omega^{-1}$. This is much higher than the free-electron-gas estimate $g_c\simeq(25\,000\ \Omega)^{-1}$. We discuss this question below in the general context of the properties of the $A15$'s and suggest three specific reasons for this effect.

It has been evident for many years that high-T_c superconductivity is almost the least unusual property of the $A15$ compounds and some other high-T_c materials. Many of them show one or more of several other, possibly related, anomalies of electronic properties, which we list.

(i) Very large phonon (T-dependent) resistivity, such that the electronic mean free path at room temperature seems to be much shorter than ϵ_F/ω_D times the lattice spacing, i.e., to violate Migdal's theorem.

(ii) Saturation of the T-dependent resistivity as T is raised at a value near the ρ_c proposed here.

(iii) Strong nonadditivity of phonon and impurity resistivity, i.e., strong violation of Matthiessen's rule, so that (again near the ρ_c proposed) the $\rho(T)$ curve is almost flat. (ii) and (iii) have been proposed already as localization phenomena by Jonson and Girvin[17] and Imry.[17]

(iv) Very large anharmonicity of lattice vibrations, as well as indications of anomalous zero-point excursion of atomic position.

(v) The well-known displacive lattice transitions.

(vi) Highly T-dependent Pauli susceptibility and peculiar specific-heat γ's.

(i), (v), and (vi) are indications of anomalously strong many-body effects due to electron-phonon coupling, effects whose characteristic energies are far greater than T_c—more similar to ω_D and ϵ_F.

It is therefore clear that conventional weak coupling BCS theory, or even the usual strong coupling theory, is inadequate to deal with these materials. We feel that, therefore, there is *a priori* reason to believe that g_c can also be strongly modified. The direction of the effect can be understood to be correct using two qualitative arguments, which may be equivalent.

First, the conductivity $\sigma(q,\omega)$ as $(q,\omega)\to 0$ is unrenormalized by many-body effects, to lowest order in scattering at least. It contains only the Fermi-surface area and mean free path, which are geometrical and hence not dynamic effects. However, the conductivity for $\omega>\omega_D$ surely is affected by phonon emission and is considerably reduced. Thus it is likely that in the early stages of scaling corresponding to the region affecting μ, g is consider-

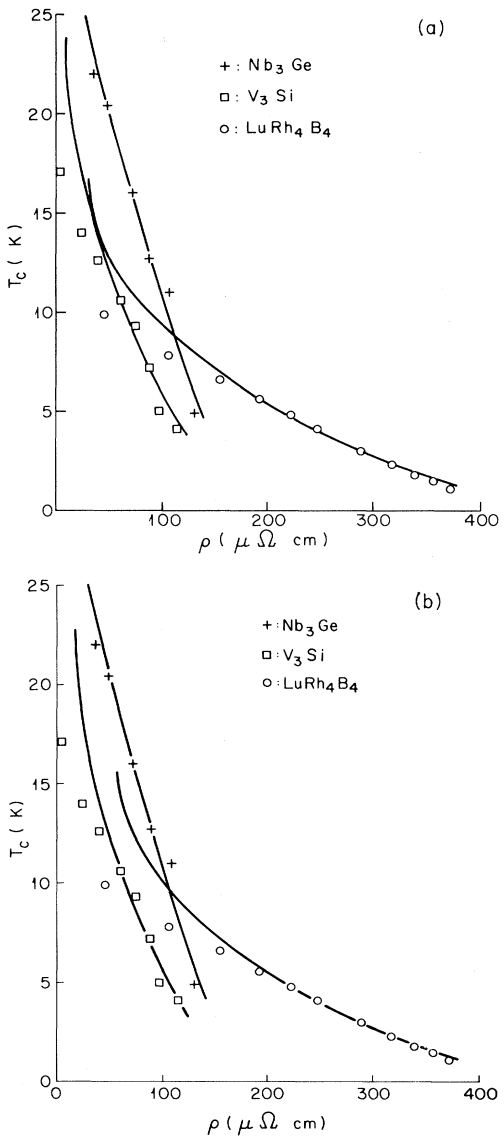

FIG. 1. Resistivity vs transition temperature of Nb_3Ge, V_3Si, and $LuRh_4B_4$. The experimental points are taken from Rowell and Dynes, Ref. 1. The solid lines are theoretical fits with (a) $\rho_c=7$, 10, and 31 $\mu\Omega$ cm for Nb_3Ge, V_3Si, and $LuRh_4B_4$, respectively, with $\ln(\epsilon_F/\omega_D)=6$, and (b) $\rho_c=13$, 18, and 57 $\mu\Omega$ cm for Nb_3Ge, V_3Si, and $LuRh_4B_4$, respectively, with $\ln(\epsilon_F/\omega_D)=5$. For both (a) and (b) we have used $\lambda=1.9$, 1.2, and 1.0, and $\omega_D=302$, 330, and 300 K for Nb_3Ge, V_3Si, and $LuRh_4B_4$, respectively. Note that the theoretical curves are not valid at small ρ.

ably smaller, and in the neighborhood of ω_D it undergoes an upward renormalization due to the removal of inelastic phonon effects. Thus the σ we see is larger than that involved in the scaling regime. In other words, estimates of the critical conductance extracted from comparisons with macroscopic values of the conductivity will be larger than the estimate obtained from the scaling regime.

Second, we have investigated the effect of a large phonon-induced attractive local interaction[16] on the scaling function $\beta(g)$ due to Hartree-type diagrams of the sort described by Altshuler *et al.*[5] for Coulomb interactions. We find a new contribution to $\beta(g)$ from phonons of the form $-\lambda_{\rm ph}/g$, i.e., a considerable strengthening of localization and hence an increase of g_c. In general, it is found that materials with a larger $\lambda_{\rm ph}$ have a larger g_c. However, our perturbation calculation is, of course, not really valid at $\lambda_{\rm ph}\sim 1$.

The compounds considered here consist of n_b orbitally degenerate tight-binding states broadened into a band by overlap. In the limit that these n_b bands are degenerate near the Fermi energy, and the local disorder does not connect states in different bands, the critical conductance increases n_b-fold (for the systems discussed $n_b=3$ or 5). If the off-diagonal two-center integrals are small, the corresponding band-mixing length scales are large, so that in both the perfect and disordered system, there is a regime of length scale where the bands are effectively degenerate, the disorder is diagonal, and g_c large by a factor n_b. At larger length scales the band mixing will be considerable, reducing g_c to its one-band value; ρ does not increase, and T_c also does not decrease. This corresponds to the observed saturation in ρ and T_c. All these arguments suggest that g_c is not a universal quantity, and although we have not shown quantitatively, it is quite plausible and expected from the above considerations that several effects may contribute, all towards the right direction, to a large renormalization of the free-electron estimate of g_c.

Finally, we note that Rosenbaum and Thomas[18] find that the critical region in σ for impurity bands seems to be anomalously large experimentally. They also find that the critical resistivity ρ_c is about an order of magnitude less than the Mott resistivity ρ_M. We conclude that localization does provide, within the framework of the theory of Abrahams *et al.*[4] a universal factor responsible for the degradation of T_c, which must be present in all materials (provided they are sufficiently disordered) over and above any contributions resulting from their specific peculiarities.[19] We suggest several reasons for g_c being renormalized.

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