Relation between the T_c degradation and the correlation gap in disordered superconductors

D. Belitz
Department of Physics and Materials Science Institute, University of Oregon, Eugene, Oregon 97403
(Received 18 November 1988; revised manuscript received 6 March 1989)

It is shown theoretically that for both bulk and thin-film superconductors the dominant contributions to the disorder-induced degradation of T_c can be expressed in terms of the disorder-induced suppression δN of the normal-state electronic density of states. This explains the correlation found experimentally between T_c and δN, and it eliminates the resistivity scale as an adjustable parameter for comparison between theory and experiment. Agreement with recent experimental results on Pb is very good. We also discuss the disorder dependence of the superconducting gap.

I. INTRODUCTION

The problem of how disorder suppresses superconductivity has been of considerable interest for some time. An important experimental step was the successful fabrication of microscopically homogeneous superconducting films.[1,2] It has become clear that the physics which governs the interplay between superconductivity and disorder in these materials is very different from the one in granular films,[3] and consequently theoretical approaches to the two different classes have taken very different routes. Early theoretical work on homogeneous materials[4] suggested that the dominant mechanism for the observed suppression of T_c is related to the mechanism suppressing the one-particle (tunneling) density of states (DOS) in the normal state.[5] This notion of a "correlation-gap mechanism"[6] for the T_c degradation was initially put forward as a separate mechanism unrelated to other disorder-generated effects which affect T_c. For instance, it had long been known that disorder increases the effective electron-phonon coupling[7] which tends to increase T_c, while a disorder-induced increase of the Coulomb pseudopotential had been shown to have the opposite effect.[8] It finally proved possible to combine all these different effects in the framework of a strong-coupling theory,[9,10] which has allowed a full microscopic description on equal footing with Eliashberg theory to come within reach. Reference 9 gave a detailed discussion of the various sources of the disorder dependence of T_c. It also showed how the previous work[4,6-8] was related to the microscopic theory, and that the latter contains all effects discussed before separately.

There have been many attempts to compare the various theoretical results with experiments. An explicit McMillan-type solution of the microscopic theory has been found.[9] Comparison with experimental results for T_c versus resistivity in bulk materials[9,11] gave good agreement. However, there are still many open questions concerning the application of the theory to experiment. For instance, the explicit solution contains a resistivity scale which is known only to within a factor of at least 2. Attempts to obtain explicit results for thin films have been plagued by the logarithmic singularities one encounters in perturbation theory in two dimensions (2D). These singularities can be remedied by various self-consistency schemes and by considering inelastic lifetime cutoffs.[10] However, realistic inelastic scattering rates in homogeneous materials are probably too small to serve the desired purpose,[12] and self-consistent schemes are somewhat arbitrary since the proper resummation of the 2D singularities is not completely understood even in the absence of superconductivity.[13] The theoretical analysis is further complicated by the fact that the films in question are in an intermediate thickness regime, and their effective dimensionalities with respect to different microscopic processes are different. Experimentally, the disorder is usually controlled by means of the film thickness, and it is not unusual for a single experiment to cover more than one dimensionality regime even with respect to one microscopic length scale. All this makes it very difficult to theoretically produce explicit results for T_c versus disorder which can be compared to experiment in a meaningful way.

On the experimental side, a clear correlation between the T_c degradation δT_c and the suppression δN of the normal-state tunneling DOS in thin films has been observed.[14] This observation lends experimental support to the idea[4,9] that the correlation-gap mechanism plays a dominant role in the suppression of T_c. However, it is also somewhat puzzling since, as already mentioned, theoretically many different sources for the T_c degradation are found, only one of which is obviously related to δN.

The purpose of this paper is to show that the theoretical relation between T_c and δN can be made precise by expressing all dominant (i.e., diffusion-related) contributions to δT_c in terms of the independent observable δN. For given clean limit parameters we thus obtain a universal relation between δT_c and δN, which can be compared with experiment without adjustable parameters. This avoids the difficulties mentioned. It allows us to apply the theory of Ref. 9 to thin films, which was not possible before, and provides a stringent and unequivocal test of the theory. Agreement with the recent experiment by

Valles *et al.*[14] is very good. A similar reasoning can be used to obtain the zero-temperature-gap parameter Δ_0. Within a two-square-well approximation we obtain satisfactory agreement with experimental results[14] for Pb, while the puzzling observations[14] on Sn apparently require a more sophisticated analysis.

II. THEORY

As a starting point I choose my recent strong-coupling theory for disordered superconductors.[9] The key observation was that in the presence of disorder the anomalous self-energy W and the renormalization function Z are not sufficient to describe the superconductor. Rather one has to keep an additional piece $Y(\varepsilon,i\omega)$ of the normal self-energy, which in the clean limit is constant and can be omitted. The frequency dependence of Y was shown to be weak, and the energy dependence was replaced by a Taylor expansion

$$Y(\varepsilon)\simeq(\varepsilon-\varepsilon^*)Y' ,$$

where $Y'=(dY/d\varepsilon)_{\varepsilon^*}$. In Ref. 9(b) ε^* was chosen to be zero. As pointed out by Valles *et al.*,[14] $\varepsilon^*=\bar{\omega}$ with an average phonon frequency $\bar{\omega}$ is a more physical choice, which we will adopt here. Reference 9(b) proceeded to calculate Y' for bulk materials as a function of resistivity, and Y' was shown to be an important source of disorder dependence of T_c. Here we instead relate Y' to the DOS $N(\bar{\omega})$. In the normal state, or well above the gap frequency, one has

$$N(\bar{\omega})=N_F\int d\varepsilon\,\delta(\bar{\omega}Z(\bar{\omega})-\varepsilon-(\varepsilon-\bar{\omega})Y') , \tag{1}$$

where N_F is the clean normal-metal DOS at the Fermi level. We thus obtain for our disorder parameter Y' the relation

$$Y'=N_F/N(\bar{\omega})-1 . \tag{2}$$

Equation (2) is independent of the dimensionality of the material and holds for arbitrary disorder.[15] It constitutes the desired relation between the parameter of the correlation-gap mechanism for the T_c degradation and an observable independent of T_c.

It is well known that Y' is not the only source of disorder dependence of T_c. Both the electron-phonon coupling[7] and the Coulomb pseudopotential[8] increase with disorder. These effects have been discussed in a unifying framework together with Y' in Ref. 9. It was shown that all disorder dependencies can be expressed in terms of correlation functions for normal conducting electrons, which in turn can be expanded in a complete set of electronic modes starting with density, current, stress, etc. Since the density is the only conserved quality in the disordered electron system, we expect the contribution of the density correlation function to be the most important because of the associated diffusion pole. Indeed this is known to be the case for both the Coulomb and the electron-phonon interaction in the limit of small disorder,[5,16] where the diffusion pole leads to nonanalyticities in the perturbation expansion, and in the limit of large disorder,[13,17] where diffusive singularities govern the critical behavior near the metal-insulator transition. We therefore employ the diffusion pole approximation (DPA) which consists in keeping only the diffusive contribution of the density correlation function to the disorder dependence of all quantities. Within DPA one has universality in the sense that the disorder only appears in the form of a wave number integral over a diffusion pole

$$\mathcal{D}(\varepsilon)=\sum_q \frac{1}{-i\varepsilon+D(q,\varepsilon)q^2} . \tag{3}$$

Here $D(q,\varepsilon)$ is a generalized electronic diffusivity, and ε is some energy variable whose meaning depends on the particular quantity under consideration. The evaluation and interpretation of terms of this form is nontrivial for 2D or quasi-2D systems, since in lowest order $D(q,\varepsilon)\equiv D^{(0)}$, and $\mathcal{D}(0)$ diverges. Within DPA, however, we can make use of the above-mentioned universality and express all disorder-dependent quantities in terms of Y'. Since Y' depends on the phonon spectrum and on the Coulomb propagator,[9] we have to specify these first. For a Debye spectrum and Thomas-Fermi screening with screening wave number κ, one obtains

$$\mathrm{Re}\mathcal{D}(0)=\frac{\pi N_F}{\mu(\zeta-3)+4\lambda}Y'\equiv\pi N_F y . \tag{4a}$$

Here λ is the usual electron-phonon coupling parameter,

$$\mu=(1/2x^2)\ln(1+x^2) \tag{4b}$$

is the Coulomb parameter[18] with $x=2k_F/\kappa$, and

$$\zeta=x\arctan x\,\ln(1+x^2) . \tag{4c}$$

For systems not too close to a metal-insulator transition, all disorder-dependent quantities can be expressed in terms of $\mathrm{Re}\mathcal{D}(0)$. Neglecting the argument of $\mathcal{D}(\varepsilon)$ has the same range of validity as neglecting the energy dependence of Y'. We find[19] a disorder-dependent electron-phonon coupling

$$\tilde{\lambda}=\lambda(1+y) , \tag{5}$$

and a disorder-dependent Coulomb pseudopotential

$$\tilde{\mu}^*=\tilde{\mu}\left[1+\frac{\tilde{\mu}}{1+Y'}(1/\mu^*-1/\mu)\right]^{-1} , \tag{6a}$$

$$\tilde{\mu}=\mu[1+(1+\zeta)y] . \tag{6b}$$

Finally we solve the strong-coupling equations[9] in two-square-well approximation, and adjust the prefactors such that in the clean limit we recover the Allen-Dynes formula,[20]

$$T_c=\frac{\omega_{\log}}{1.2}\exp\left[\frac{-1.04(1+\tilde{\lambda}+Y')}{\tilde{\lambda}-\tilde{\mu}^*[1+0.62\tilde{\lambda}/(1+Y')]}\right] . \tag{7}$$

It is easy to see that within DPA $\omega_{\log}$ does not depend on Y'. Equation (2) and (4)–(7) are the desired relation between T_c and the tunneling DOS. A similar idea of expressing δT_c in terms of δn (for a different theory for δT_c) was formulated earlier by Muttalib.[21]

Let us also consider the disorder dependence of the gap parameter Δ_0 by solving the strong-coupling equations of Ref. 9(b) at zero temperature in two-square-well approximation. We find $\Delta_0/\omega_0 = u$ where ω_0 is the maximum phonon frequency, and u is the solution of the transcendental equation

$$u = 2a \exp\left[-\frac{1.04[1+\tilde{\lambda}F(u)+Y']}{\tilde{\lambda} f_\lambda(u) - \tilde{\mu}^*[1+f_\mu \tilde{\lambda}/(1+Y')]} \right] . \quad (8)$$

Here

$$f_\lambda(u) = \frac{2}{\ln(2/u)} \int_u^1 \frac{dx}{(x^2-u^2)^{1/2}} \alpha^2 f(x) , \quad (9a)$$

$$f_\mu = 2\int_1^\infty dx\, \alpha^2 f(x)/x , \quad (9b)$$

with

$$\alpha^2 f(y) = \frac{1}{\lambda} \int_0^\infty dx\, \alpha^2 F(x\omega_0)/(x+y) , \quad (9c)$$

and

$$F(u) = 2\int_u^\infty dx\, f(x) + 2\int_u^1 \frac{dx}{(x^2-u^2)^{1/2}} x f(x) , \quad (10a)$$

with

$$f(y) = (\omega_0/\lambda) \int_0^1 dx\, \alpha^2 F(x\omega_0)/(x+y)^2 . \quad (10b)$$

a in Eq. (8) is a phenomenological factor similar to the prefactor 1/1.2 in Eq. (7), which is needed since the two-square-well approximation is known[20] to give an incorrect prefactor. A choice of $a = 1.4$ with a Debye spectrum gives very reasonable values for $2\Delta_0/T_c$ in the clean limit ranging from 3.5 for $\lambda = 0.5$ to 4.8 for $\lambda = 2.0$. This shows that T_c and Δ_0 have been treated consistently.

III. DISCUSSION

Equation (7) has been compared with experiments on Pb in Ref. 14, so we can confine ourselves to a brief discussion of the clean limit parameters entering Eq. (7). It is well known that if one starts to make a clean superconductor disordered, there often is an initial sharp drop in T_c. This is connected with the smearing of anisotropies in both the electron and the phonon system, and it gives way to a more gradual behavior once the "dirty limit" has been reached.[22] In the experimental results of Refs. 2 and 14 the initial sharp drop is apparent, and the dirty limit has clearly been reached at $R \simeq 100\Omega/\square$. The present theory is concerned with the gradual behavior in the dirty limit and is not designed to deal with how the dirty limit is reached. $T_c(Y'=0)$ according to Eq. (7) therefore has to be interpreted as the T_c of a hypothetical Pb which stays isotropic all the way to the clean limit. This T_c will be smaller than that of real clean Pb. It is interesting to note that Allen and Dynes[20] obtain tunneling values for λ, $\omega_{\log}$, and μ^* which lead to $T_c = 6.49$ K which is consistent with a linear extrapolation from the dirty limit of the data in Refs. 2 and 14. If we use Eq. (7) with the values for λ, $\omega_{\log}$, and μ^* taken from Ref. 20 we obtain the curve shown in Fig. 1. Over the range of Y' considered, this curve is an almost exact parallel displacement of the one obtained in Ref. 14 with a larger value of λ which gives the actual clean limit T_c.

The Y' dependence of the gap parameter resulting from Eq. (8) with a Debye spectrum is shown as the solid line in Fig. 1. $2\Delta_0/T_c$ decreases slightly, while in experiment it stays constant, though the data scatter considerably. It should be noticed, however, that the disorder dependence of Δ_0 as given by Eq. (8), and therefore also that of $2\Delta_0/T_c$, depends on the form of the Eliashberg function. Inspection shows that an α^2F with more spectral weight at large frequencies produces a weaker decrease of Δ_0/T_c with increasing Y'. The dashed line in Fig. 1 shows the result one gets for an Einstein spectrum with $a = 2.3$, which fits the data very well. However, only a McMillan inversion procedure applied to the equations of Ref. 9 will show if this is not an artifact of the two-square-well approximation. We conclude that in the case of Pb there is excellent agreement between theory and experiment for the T_c degradation, and reasonable agreement for Δ_0/T_c, though the latter requires more theoretical work.

For Sn the situation is less satisfactory. Experiment[14] shows that amorphous Sn is strongly coupled, and for $R \geq 250\Omega/\square$ the behavior of T_c is very similar to that of Pb. However, with increasing disorder the ratio $2\Delta_0/T_c$ drops from 4.5 at small Y' to values less than 3.5 at $Y' \cong 0.8$. The sensitivity of the Y' dependence of Δ_0 to the form of the Eliashberg function which was previously demonstrated does lead to a stronger decrease of Δ_0/T_c with increasing Y' if α^2F has more spectral weight at small frequencies. However, we found it hard to obtain a drop as rapid as the one observed in experiment. Again it will take a McMillan inversion procedure applied to the

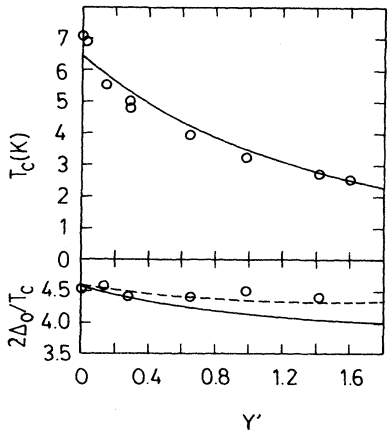

FIG. 1. Solid curves are theoretical results for the transition temperature and the ratio $2\Delta_0/k_BT_c$. A Debye spectrum was used and $\lambda = 1.55$, $\mu^* = 0.105$, $\omega_{\log} = 56$ K. The dashed curve is the theoretical result for $2\Delta_0/k_BT_c$ with an Einstein spectrum. Circles are experimental results for Pb from Ref. 14.

equations of Ref. 9 rather than the simple two-square-well approximation to decide if the present theory can explain these observations. The full solution will also be necessary to see if the presence of Y' remedies the convergence problems which were encountered in an attempt to analyze these materials with Eliashberg theory.[2] These questions are left for future investigations.

ACKNOWLEDGMENTS

I am much indebted to R. C. Dynes and J. M. Valles, Jr. for making their data available prior to publication and for helpful correspondence. This work was supported by the Materials Science Institute at the University of Oregon.

[1]J. M. Graybeal and M. A. Beasley, Phys. Rev. B **29**, 4167 (1984).
[2]R. C. Dynes, A. E. White, J. M. Graybeal, and J. P. Garno, Phys. Rev. Lett. **57**, 2195 (1986).
[3]H. M. Jaeger, D. B. Haviland, A. M. Goldman, and B. G. Orr, Phys. Rev. B **34**, 4920 (1986).
[4]H. Fukuyama, H. Ebisawa, and S. Maekawa, J. Phys. Soc. Jpn. **53**, 1919 (1984); S. Maekawa, H. Ebisawa, and H. Fukuyama, *ibid.* **53**, 3560 (1984).
[5]B. L. Altshuler and A. G. Aronov, Solid State Commun. **30**, 115 (1979); B. L. Altshuler, A. G. Aronov, and P. A. Lee, Phys. Rev. Lett. **44**, 1288 (1980).
[6]D. Belitz, J. Phys. F **15**, 2315 (1985).
[7]B. Keck and A. Schmid, J. Low Temp. Phys. **24**, 611 (1976).
[8]P. W. Anderson, K. A. Muttalib, and T. V. Ramakrishnan, Phys. Rev. B **28**, 117 (1983).
[9](a) D. Belitz, Phys. Rev. B **35**, 1636 (1987); (b) D. Belitz, *ibid.* **35**, 1651 (1987).
[10]D. A. Browne, K. Levin, and K. A. Muttalib, Phys. Rev. Lett. **58**, 156 (1987).
[11]D. Belitz, Phys. Rev. B **36**, 47 (1987).
[12]D. Belitz, S. De Souza-Machado, T. P. Deveraux, and D. W. Hoard, Phys. Rev. B **39**, 2072 (1989); T. P. Deveraux and D. Belitz (unpublished).
[13]A. M. Finkelshtein, Z. Phys. B **56**, 189 (1984); C. Castellani, G. Kotliar, and P. A. Lee, Phys. Rev. Lett. **59**, 323 (1987).
[14]J. M. Valles, Jr., R. C. Dynes, and J. P. Garno (unpublished); R. C. Dynes and J. M. Valles, Jr. (private communication).
[15]We note, however, that at very large disorder, as the material approaches a metal-insulator transition, Y' will develop a strong ε dependence, and the first-order Taylor expansion leading to Eq. (2) will no longer be a good approximation.
[16]A. Houghton and H. Won, J. Phys. C **18**, 2507 (1985); T. R. Kirkpatrick and D. Belitz, Phys. Rev. B **34**, 2168 (1986).
[17]T. R. Kirkpatrick and D. Belitz, Phys. Rev. B **34**, 9008 (1986); C. Castellani and G. Kotliar, *ibid.* **34**, 9012 (1986).
[18]P. Morel and P. W. Anderson, Phys. Rev. **125**, 1263 (1962).
[19]Since Ref. 9(b) also used the DPA, Eqs. (5) and (6) can simply be obtained by eliminating the resistivity from Eqs. (2.29) and (2.30) of Ref. 9(b) and using Eq. (2). The nontrivial point is that the result is valid for 2D as well as for 3D systems.
[20]P. B. Allen and R. C. Dynes, Phys. Rev. B **12**, 905 (1975).
[21]K. A. Muttalib, Phys. Rev. B **27**, 4628 (1983).
[22]P. W. Anderson, J. Phys. Chem. Solids **11**, 26 (1959).