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Estimate of density-of-states changes with disorder in A-15 superconductors*

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Data are presented which show that the density of states changes dramatically, as does T_c, in both Nb_3Sn and Nb_3Ge samples which were disordered by α-particle irradiation. The density of states is derived from the strong-coupling modifications to the theory of type-II superconduction along with measurements of $(dH_{c2}/dT)_{Tc}$ and the residual normal-state resistance ρ_0.

I. INTRODUCTION

Various models[1] have been proposed for the rapid depression of T_c with disorder found in A-15 superconductors, and also in other d-band superconductors such as Nb.[2-4] The behavior of the density of states and its relationship of T_c and disorder is crucial to this problem.

In this paper we use a thermodynamic method to estimate changes in the density of states N as a function of disorder in A-15 materials. Although there are some possible problems with the absolute accuracy of the method, changes in N of the order of a factor of 4 are observed as T_c changes from near 20 K to about 4 K, and the method nicely demonstrates these large changes in N.

As far as we know, there is no actual theory for how disorder affects the density of states. This question was considered by Crow *et al.*[2] with regard to drastic changes in the T_c of transition-metal superconductors when they were made very disordered by deposition onto cryogenic substrates, and there has been further discussion recently for A-15 superconductors by Dynes and Varma.[4] In the paper of Crow *et al.*,[2] a plausibility argument was given for density-of-states smearing by use of the uncertainty relationship

$$\Delta E \Delta t \sim \hbar . \tag{1}$$

For mean free paths l of the order of interatomic spacings, it is reasonable to expect significant smearing. We will consider this problem in more detail later. In the case of the A-15 superconductors where extremely sharp structure is expected in the density of states near the Fermi level, a significant change with l can be expected. Since T_c is expected to correlate with N,[2,4] changes with T_c are expected with decreased N due to a decreased l. The measurements presented here show this effect. We emphasize that this approach based on N decreasing with l, implies that the effects due to defects are additive and are reflected in the normal-state resistivity ρ_0. The detailed nature of the defect is relatively unimportant except for the change in ρ_0 which determines the effect on N.[5]

II. METHOD

To estimate changes in N with changes in ρ_0, we first consider a phenomenological analysis for N in terms of $(dH_{c2}/dT)_{T_c}$ and ρ_0. We caution that this analysis does not adequately account for strong coupling, as will be made clear in Sec. III. At 0 K, one can equate the gap energy from the BCS theory to the condensation energy

$$\tfrac{1}{2}N^*\Delta^2 = H_c^2(0)/8\pi , \tag{2}$$

where N^* is the specific-heat density of states. $H_c(0)$ can then be related to $H_{c2}(T)$. Near T_c, we have

$$H_{c2} = \sqrt{2}\,\kappa H_c , \tag{3}$$

and if the Ginzburg-Landau parameter κ is slowly varying, we get

$$H'_{c2}(T_c) \equiv -\left(\frac{dH_{c2}}{dT}\right)_{T_c} = -\sqrt{2}\,\kappa\left(\frac{dH_c}{dT}\right)_{T_c} .$$

Using the BCS relation $H_c(T) = 1.74 H_c(0)(1 - T/T_c)$, we find

$$\left(\frac{dH_{c2}}{dT}\right)_{T_c} = -\sqrt{2}\,\kappa 1.74\,\frac{H_c(0)}{T_c} \tag{4}$$

if κ is slowly varying. Assuming $\Delta = \alpha T_c k_B$, i.e., α depends on the coupling, then the density of states can be expressed as

$$N^* = \frac{(dH_{c2}/dT)^2_{T_c}}{\kappa^2(1.74)^2 k_B^2 \alpha^2 8\pi}\;\frac{\text{erg}}{\text{cm}^3\,\text{K}} , \tag{5}$$

where κ is given by

$$\kappa = 0.96\,\lambda_L/\xi_0 + 0.7\,\lambda_L/l ,$$

where λ_L is the Landau penetration depth, ξ_0 the coherence length, and l the electronic mean free path, or in terms of the resistivity ρ_0 and γ,

$$\kappa \sim 7.53\times 10^3 \rho_0 \gamma^{1/2}(1 + 1.3 l/\xi_0) . \tag{6}$$

γ is the specific-heat coefficient and is related to the density of states through $\gamma = \frac{2}{3}\pi^2 k_B^2 N(0)^*$. Then,

$$N^* = [H_c'(T_c)]^2[\alpha^2(1.74)^2 k_B^2 8\pi\gamma \times (7.53\times 10^3\rho_0)^2(1+1.3l/\xi_0)^2]^{-1}$$

and

$$\gamma = 2.16\times 10^{-5} H_{c2}'(T_c)\,\frac{1}{\rho_0(1+1.3l/\xi_0)}\,\frac{\text{erg}}{\text{cm}^3\,\text{K}^2},$$

which can be written

$$\gamma = -6.5\times 10^{-13}\left(\frac{dH_{c2}}{dT}\right)_{T_c}\frac{a_0^3}{\rho_0}\,\frac{1}{1+1.3l/\xi_0}\,\frac{\text{J}}{\text{mol K}^2}, \quad (7)$$

where a_0 is the lattice spaceing in Å. Hence, by measuring $H_{c2}'(T_c)$ and dividing by ρ_0 one can essentially get γ. The $(1+1.3l/\xi_0)$ correction becomes important when $l \sim \xi_0$.

A better estimate of γ in the strong-coupling regime can be obtained by going to the microscopic theory. In the dirty limit, one can write

$$H_{c2}'(T_c) = (4/\pi)k_B c/eD,$$

where $D = \frac{1}{3}v_F l$ is the diffusion constant, v_F is the Fermi velocity, and c is the velocity of light. Rainer and Bergmann[6] then show that in the strong coupling regime $H_{c2}'(T_c)$ can be written

$$H_{c2}'(T_c) = 1.27\eta c k_B/eD^*, \quad (8)$$

where η is a strong-coupling correction that depends on $T_c/\langle\omega\rangle$. For $T_c/\langle\omega\rangle \sim 0.1$, then $\eta \sim 1.25$ and it goes to one as $T_c/\langle\omega\rangle$ goes to zero. (Note that in their paper the factor eD^* is inverted.) The diffusion constant D^* is defined as $D^* = D/(1+\lambda)$, where λ is the electron-phonon coupling coefficient. Then using the single-spin density of states and the expression for $\rho = [2e^2N(0)D]^{-1}$, we can write

$$H_{c2}'(T_c) = 1.27\eta e N\rho_0(1+\lambda)ck_B 2.$$

Hence, in terms of H_{c2}', the electronic specific heat can be expressed as

$$\gamma = \tfrac{2}{3}\pi^2 N^* k_B^2 = [\pi^2/3(1.27)]H_{c2}'k_B/\rho_0\eta ec$$
$$= 2.2\times 10^{-5}(H_{c2}'/\eta\rho_0)\,\text{erg/cm}^3\,\text{K}^2,$$

where ρ_0 is in Ω cm, and finally we get

$$\gamma = 6.5\times 10^{-13} H_{c2}'(T_c)(a_0^3/\rho_0\eta)(\text{J/mol K}^2). \quad (9)$$

If $l \gtrsim \xi_0$ then we suppose the expression will have the $(1+1.3l/\xi_0)$ factor found in the phenomenological analysis and hence

$$\gamma = 6.5\times 10^{-13} H_{c2}' a_0^3\left[\rho_0\eta\left(1+\frac{1.3l}{\xi_0^*}\right)\right]^{-1}\frac{\text{J}}{\text{mol K}^2}, \quad (10)$$

where now ξ_0^* is $\xi_0/(1+\lambda)$.

III. EXPERIMENTAL

The samples used in this study were Nb_3Ge and Nb_3Sn films prepared by electron-beam codeposition. Damage was provided by irradiating the samples with 2.5-MeV α particles. After a relatively heavy irradiation, where T_c was depressed to the order of 4 K, both ρ_0 and $(dH_{c2}/dT)_{T_c}$ were determined. $(dH_{c2}/dT)_{T_c}$ was determined by measuring the critical field as function of T near T_c in a 5-T superconducting magnet, with the sample gauge length normal to the field. To change T_c, the samples were annealed at various temperatures. At each new T_c, $(dH_{c2}/dT)_{T_c}$ and ρ_0 were also measured.

IV. DATA

In Fig. 1, we show plots of $(dH_{c2}/dT)_{T_c}/\rho_0$ vs T_c for both Nb_3Ge and Nb_3Sn films. In Figs. 2 and 3, we show the variation of $H_{c2}'(T_c)$ and ρ_0 for each T_c reached after annealing. It is evident that H_{c2}' is slowly varying, with most of the changes in $H_{c2}'(T_c)/\rho_0$ coming from the larger changes in ρ_0. The fact that dH_{c2}/dT is slowly varying as T_c decreases also implies that $H_{c2}(0)$ decreases since by Eqs. (3) and (4) it can be seen that $H_{c2}(0)$ is proportional to the product of $H_{c2}'(T_c)$ and T_c. Note that in the extreme dirty limit, γ is related to $H_{c2}'(T_c)/\rho_0$, since l/ξ_0 becomes small. However, in the case of Nb_3Sn, where for clean films $l \gtrsim \xi_0^*$, and even in the case of Nb_3Ge, where in the cleanest films $l \sim 5\xi_0^*$ one must include the $(1+1.3l/\xi_0^*)$ correction. An estimate of l can be obtained from ρ_0 and various estimates can be made of ξ_0^*. We assume that for $\rho_0 \sim 150\ \mu\Omega$ cm,

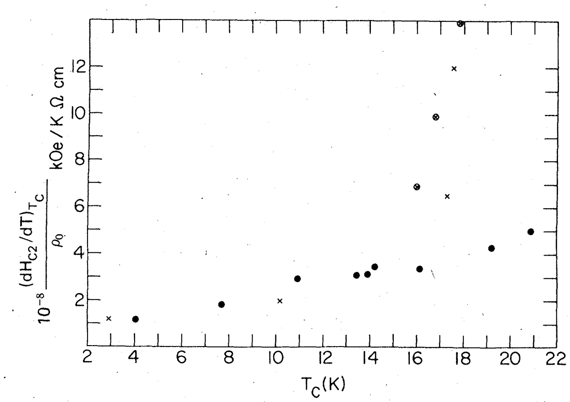

FIG. 1. Nb_3Ge–● is represented by three sets of data. In each case initially high-T_c samples, 19–21 K, were measured, radiation damaged, and recovered by subsequent annealing. The Nb_3Sn–x data was acquired in the same manner except for three Nb_3Sn–⊗ samples which were only measured "as grown."

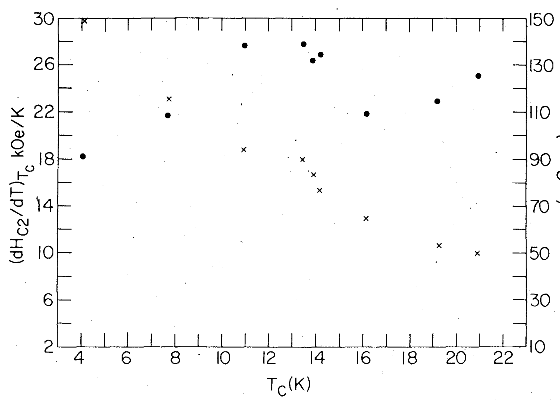

FIG. 2. Raw data for Nb_3Ge used to construct Fig. 1. $(dH_{c2}/dT)_{T_c}$ –● is seen to be relatively constant with most of the variation in ρ_0–x. The complimentary points for each sample are vertically opposite each other.

$l \sim 5$ Å, and thus, $l\rho_0 \sim 750$ Å $\mu\Omega$ cm. In Fig. 4, we show $\gamma\eta$ for Nb_3Sn and Nb_3Ge assuming that $\xi_0^* \sim 50$ Å in Eq. (10)

V. DISCUSSION

The essential philosophy in presenting these data is not to get accurate numerical values of γ or N since there are some problems with the method which will be discussed below. The main point we wish to emphasize is the large change in γ with the transition temperature of the superconductor. It can be seen that in a qualitative way γ depends linearly on T_c. Excluding the regions where ρ_0 is very small, i.e., very long l, or where ρ_0 is very large, i.e., N and T_c are saturated, it can be seen that since $H'_{c2}(T_c)$ is slowly varying, γ goes approximately as $1/\rho_0$. This can be understood on the basis of a simple model which emphasizes the smearing of the density of states as l gets smaller.

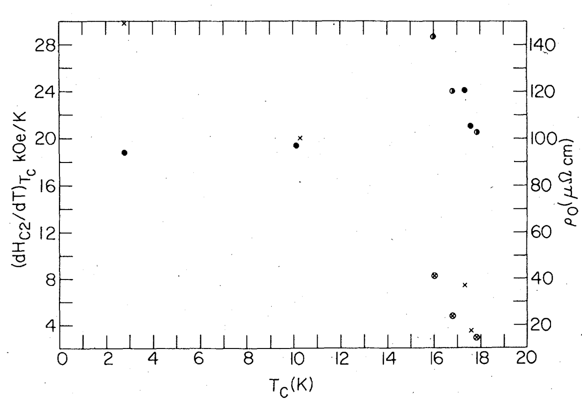

FIG. 3. Raw data for Nb_3Sn used to construct Fig. 1. $(dH_{c2}/dT)_{T_c}$ –● is relatively constant compared to variation in ρ_0–x. The complimentary points for each sample are vertically opposite each other. ◑ represents $(dH_{c2}/dT)_{T_c}$ for "as grown" samples and ⊗ represents corresponding values of ρ_0.

Consider a sharp structure in the density of states near the Fermi level E_F with a width of about 100 K. It is argued that N will decrease if the uncertainty in the energy ΔE approaches about 100 K. From the uncertainty relationship, a length l can be derived from

$$\Delta E \Delta t = \Delta E = l/v_F = \hbar$$

for $\Delta E = 100$ K and $v_F = 10^7$ cm/sec, $l \sim 100$ Å. Hence, when l is of the order of 100Å, smearing of N can be expected. Since ΔE goes as $1/l$, it is also true that $\Delta E \propto \rho_0$. If one assumes a constant number of states in the density-of-states peak near E_F, then the area of the curve becomes $N\Delta E$ for small l. Since $\Delta E \propto \rho_0$, we have $N \propto 1/\rho_0$. Of course, this analysis is only qualitative in nature and no attempt has been made to consider $N^*(0) = (1+\lambda)N(0)$.

It should be mentioned that the relatively large initial changes of γ with T_c shown in Fig. 4 for the case of Nb_3Sn might be explained by the recent argument of Nettle and Thomas,[7] which indicates that in the theory of superconductivity the peak density of states at the Fermi level must be averaged over energies of the order of the phonon cutoff, and T_c would depend on this average N, i.e., $\bar{N}$. We would then expect γ which goes as $N^*(0) = N(0)(1+\lambda)$ to change more rapidly with disorder than T_c which is proportional to $\bar{N}$, since $N(0)$ is a sharper function than $\bar{N}$ and would be more easily affected by small amounts of disorder.

It is of some interest to calculate the value of γ at T_{c0} for both Nb_3Sn and Nb_3Ge. We find in Nb_3Ge that at 20 K, $\gamma\eta \sim 35$ (mJ/mol K^2), or $\gamma = 28$

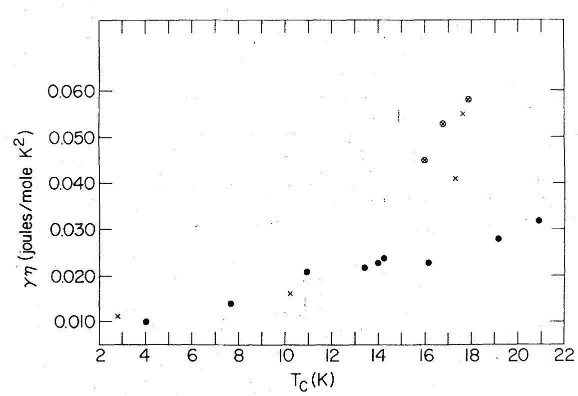

FIG. 4. $\gamma\eta$ is shown for Nb_3Ge–● and Nb_3Sn–x, ⊗ according to Eq. (10).

(mJ/mol K^2). In the case of Nb_3Sn which is *not* in the dirty limit, the values are more uncertain. If we assume $\xi_0^* \sim 50$ Å, then for $T_c = 17.8$, $\gamma\eta \sim 60$ (mJ/mol K^2) or $\gamma \sim 48$ (mJ/mol K^2). The specific-heat values are[8] ~52 (mJ/mol K^2) for Nb_3Sn and[9] ~24 (mJ/mol K^2) for Nb_3Ge. We think this agreement is reasonable. In the clean limit, the problem is very difficult since ξ_0^* is not really known.

We also briefly mention that there are some other problems with getting γ from $H'_{c2}(T_c)$. In the regime where $l \sim a$, and there is the possibility of saturation in the resistivity, it is not clear how H_{c2} is related to l and the resistivity. Finally, it has been assumed there is no paramagnetic limiting, which is expected to make only a very small error in $H'_{c2}(T_c)$.[10]

VI. SUMMARY

Results are presented which show that the density of states decreases in A-15 samples with different T_c's owing to different states of disorder. This change is quite sizable and appears to be about a factor of four from the undamaged T_{c0} to the lowest T_c caused by α-particle damage. It can also be seen that to first order, the density of states goes inversely as the residual resistance. This view leads one to believe that any "defect" which affects the scattering time and ρ_0 will change N and T_c.

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[1]A. R. Sweedler, D. G. Schweitzer, and G. W. Webb, Phys. Rev. Lett. 33, 168 (1974); J. M. Poate, L. R. Testardi, A. R. Storm, and W. M. Augustyniak, *ibid.* 35, 1290 (1975); A. E. Karkin, V. E. Arkhipov, B. N. Goshchitskii, E. P. Romanov, and S. K. Sidorov, Phys. Status Solidi A 38, 433 (1976).
[2]J. E. Crow, M. Strongin, R. S. Thompson, and O. F. Kammerer, Phys. Lett. 30, 161 (1969).
[3]M. M. Collver and R. H. Hammond, Phys. Rev. Lett. 30, 92 (1973).
[4]R. C. Dynes and C. M. Varma, J. Phys. F 6, L215 (1976).
[5]H. Lutz, H. Weismann, O. F. Kammerer, and M. Strongin, Phys. Rev. Lett. 36, 1576 (1976); C. M. Varma and R. C. Dynes, in *Superconductivity in d- and f- Band Metal,* edited by D. H. Douglass (Plenum, New York, 1976).
[6]D. Rainer and G. Bergmann, J. Low Temp. Phys. 14, 501 (1974).
[7]S. J. Nettle and H. Thomas, Solid State Commun. 21, 683 (1977).
[8]L. J. Vieland and A. W. Wicklund, Phys. Rev. 166, 424 (1968).
[9]J. M. E. Harper, Ph.D. thesis (Stanford University, 1975) (unpublished).
[10]D. Saint-James, E. J. Thomas, and G. Sarma, *Type II Superconductivity* (Pergamon, New York, 1969).