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Effects of disorder on properties of $A15$ materials

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We have calculated the effects of disorder on the density of states, Fermi velocity, and Drude plasma frequency for V and its $A15$ compounds V_3X, with $X=$Al, Ga, Ge, Si, and Sn, and for Nb and its $A15$ compounds Nb_3X, with $X=$Al, Ga, Ge, Si, Sn, and Sb using the electron-lifetime model and the results of band-structure calculations. In most cases the density of states and the superconducting transition temperature T_c are found to decrease with increasing disorder, in qualitative agreement with experiment. Exceptions are Nb_3Sb and Nb_3Si, for which we have found a small increase in T_c. We are also presenting calculations of the effects of disorder on the mean free path, BCS coherence length, London penetration depth, Ginzburg-Landau κ, and the temperature dependence of the upper critical field for the above materials. Comparison with the exisiting experimental data is made.

I. INTRODUCTION

The $A15$ compounds include materials having the highest superconducting transition temperature (T_c) known. Many of the $A15$'s also have very high critical fields and exhibit unusual properties for the electrical resistivity, magnetic susceptibility, and Knight shifts. As a result of the above properties and the desire to obtain even higher T_c and critical fields, these compounds have been extensively studied[1] both theoretically and experimentally over the past 25 years.

Irradiation[2] of superconducting materials has recently become a very active area of research due to the use of superconducting magnets in high-field applications. In applications such as bending magnets for beams in high-energy particle accelerators and magnetic confinement in proposed fusion reactors where the magnets must operate in an irradiation environment, the knowledge of the response of the critical properties of the superconductors to that irradiation must be known. Disorder induced by crystal growth conditions, α-particle irradiation, and neutron damage yields a remarkable depression of T_c for the high-temperature superconducting $A15$ compounds Nb_3Sn, V_3Si, and similar materials. Testardi *et al.*[3] have pointed out the universal nature of the depression of T_c by correlating it with the residual resistance ratio for various types of damage. A thorough investigation of neutron irradiation by Sweedler *et al.*[2] demonstrated that the relative decrease of T_c in many $A15$ compounds followed a similar pattern as a function of the neutron flux. By contrast, disorder appears to have little influence on Nb (Ref. 3) that remains superconducting to $T_c \approx 9$ K over a wide range of residual resistance. Further complication is provided by the disorder-enhanced superconducting temperature (from 1 to 5 K) in Mo_3Ge (Ref. 4) which also exhibits the $A15$ structure.

The nature of the defects responsible for the depression of the superconducting temperature is not clear. On one hand, site-exchange disorder (replacement of A atoms by B atoms and vice versa in the A_3B structure) has been identified as an important defect in diffraction studies.[2] On the other hand, an "unknown defect," perhaps in the form of atom displacement, which may be associated with the loss of translational symmetry has been suggested as a primary cause for the observable effects.[3]

The effect of static disorder on the critical temperature T_c has been studied extensively in $A15$ compounds.[5–8] However, the mechanism of the reduction of T_c in high-T_c $A15$ superconductors has remained a source of controversy. On the basis of the phonon-exchange mechanism of the superconducting electron pairing, it is natural to explore changes in the electron density of states (DOS) which may have profound influence on the transition temperature. This connection has been explored for a sharp DOS in a one-dimensional- (1D) type structure, which is easily distorted by disor-

der.[5] Unfortunately, there is a great deal of uncertainty as to how the energy smearing of the DOS, $N(E)$, will take place. The only microscopic theory is due to Labbé and Van Reuth[5] but it is based on a special type of disorder and an oversimplified 1D model of an $A15$ compound. Some other explanations have been based on the theory of dirty superconductors with anisotropic gaps.[6] However, as Gurvitch *et al.*[6] pointed out, the anisotropy necessary for explaining this reduction is unreasonably large. Appel[7] has discussed the effect of long-range order on high T_c. Another point of view has focused on the acoustic-plasmon mechanism,[8] some arguments have been based on general smearing of $N(E)$ with disorder,[9] and finally a microscopic treatment[10] of the consequences of both static and thermal disorder for superconductivity for a nonconstant DOS was presented using the generalized Eliashberg gap equations.

The purpose of the present work is, by using the electron lifetime model introduced by Testardi and Mattheiss,[9] to calculate $N(E)$, the Fermi velocity $v_F(E)$, the Drude plasma frequency $\Omega_p(E)$, the mean free path l, the $T=0$ BCS coherence length ξ_0, the $T=0$ London penetration length λ_L, the Ginzburg-Landau κ near T_c, and the temperature dependence of the upper citical field near T_c as a function of disorder broadening Γ (or residual resistivity ρ_0) for V, Nb, and the $A15$ compounds Nb_3X, V_3X, with $X=$Al, Ga, Ge, Si, Sn, and Nb_3Sb. In applying this scheme, we have used the quantities $N(E)$, $v_F(E)$, and $\Omega_p(E)$ of the perfectly ordered materials derived from the band-structure calculations of Boyer *et al.*[11] for V and Nb and of Klein *et al.*[12,13] for the $A15$ compounds. We wish to emphasize that a good quantitative understanding[13] of the superconducting properties of the fully stoichiometric $A15$ compounds has been derived from these band-structure results without additional assumptions regarding quasi-one-dimensionality or resorting to model DOS singularities. A preliminary report of the present work has been given elsewhere.[14]

II. ELECTRON LIFETIME MODEL

As already discussed, the nature of the defect or defects responsible for the properties of $A15$ materials is not clear. Testardi and Mattheiss[9] assumed that the defects (without specifying their exact nature) just broaden the DOS via the electron relaxation time τ, which decreases with disorder. If $F(E,\Gamma=0)$ is a quantity corresponding to the perfectly ordered material whose energy dependence near the Fermi energy is known [here $F(E,\Gamma=0)$ stands for $N(E)$, $v_F(E)$, or $\Omega_p(E)$], we can take the disorder effects into account with the convolution

$$F(E,\Gamma)=\int S(E,E',\Gamma)F(E',\Gamma=0)dE' , \quad (1)$$

where $S(E,E',\Gamma)$ is a broadening function that depends upon the electron damping $\Gamma=\hbar/\tau$. The exact form of the broadening function is not too crucial when the broadening is large compared to any fine structure in $F(E,\Gamma=0)$. All the calculations given below were done with a Lorentzian of half-width $\Gamma=\hbar/\tau$ as a broadening function. The exact form is

$$S(E,E',\Gamma)=\pi^{-1}\frac{\Gamma}{(E-E')^2+\Gamma^2} . \quad (2)$$

The electron lifetime τ can be estimated from the Drude plasma frequency Ω_p using the standard residual resistivity formula

$$\rho_0=4\pi\hbar\frac{\Gamma}{\hbar^2\Omega_p^2}=\frac{4\pi}{\Omega_p^2\tau} . \quad (3)$$

Using this procedure, Testardi and Mattheiss[9] calculated $N(E)$, $\Omega_p(E)$, and $v_F(E)$ as a function of Γ or ρ_0 for V, Nb, V_3Si, Nb_3Ge, and Nb_3Sn. Further assuming that the electron-phonon parameter $\lambda\sim N(E_F)$, they found good agreement with experiment for the depression of T_c with disorder for V_3Si and Nb_3Sn. We have made all the calculations given below with a Lorentzian broadening function while Testardi and Mattheiss[9] used a thermal broadening function. To avoid complications from the Lorentzian wings in determining E_F for the high values of Γ, one has to integrate the $N(E,\Gamma)$ starting from much more negative values of E than for the $\Gamma=0$ case. We have also normalized Eq. (2) with $\int S(E,E',\Gamma)\,dE'$ since we only do the E' integration on a finite-energy grid, and the integration of the Lorentzian is not exactly equal to one. Of course, one must also be careful to keep the total number of states Q in the energy band constant as we increase the electron width Γ. The Fermi energy $E_F(\Gamma)$ in the disordered state is determined from the constraint that the total number of states Q is constant, i.e.,

$$Q=\int_{-\infty}^{E_F(\Gamma=0)}N(E,\Gamma=0)\,dE=\int_{-\infty}^{E_F(\Gamma)}N(E,\Gamma)\,dE . \quad (4)$$

This is of course true for a defect that preserves the number of electrons in the crystal, e.g., disorder, as in our case, but is not true for, say, interstitials or vacancies. We have found for all the cases we examined, that $E_F(\Gamma)$ increases with disorder.

III. RESULTS AND DISCUSSION

Using Eqs. (2) and (3) and the band-structure results[11,12] of the perfectly ordered $A15$ compounds we have calculated $N(E_F,\Gamma)$, $v_F(E_F,\Gamma)$, and $\Omega_p(E_F,\Gamma)$ as a function of electron damping Γ, which is a measure of the disorder. Figures 1—3 show the calculated $N(E_F,\Gamma)$, $v_F(E_F,\Gamma)$, and $\Omega_p(E_F,\Gamma)$ as a function of electron width Γ. To obtain $N(E_F)$, v_F, and Ω_p as a function of residual resistivity ρ_0 or broadening temperature $T_B=\Gamma/k_B$ (k_B is Boltzmann's constant), one has to use the formulas that relate ρ_0 to Γ and to T_B. In particular, we have Γ given by

$$\Gamma=\frac{\hbar}{\tau}=1.34\times10^{-4}\Omega_p^2\rho_0\ , \tag{5}$$

the broadening temperature T_B given by

$$T_B=1.554\Omega_p^2\rho_0\ , \tag{6}$$

and the mean free path l given by

$$l=v_F\tau=4.91\times10^4 v_F/\Omega_p^2\rho_0\ , \tag{7}$$

where v_F is in 10^8 cm/sec, l in Å, ρ_0 in $\mu\Omega$ cm and $\hbar\Omega_p$ and Γ in eV. Equation (5) can be used to express $N(E_F)$, Ω_p, and v_F as a function of ρ_0.

We wish to point out that the information on Figs. 1—3 is enough to calculate, with the help of Eqs. (5)—(7), various properties (mean free path, $T=0$ BCS coherence length, London penetration depth at Ginzburg-Landau κ near T_c, and the temperature dependence of the upper critical field near T_c) of these materials as a function of ρ_0. In particular, we have that the BCS coherence length at 0 K is given by[9]

$$\xi_0=\frac{1.38\times10^4 v_F}{(1+\lambda)T_c}\ . \tag{8}$$

The London penetration depth at 0 K is given by

$$\lambda_L=\frac{1.98\times10^3(1+\lambda)^{1/2}}{\Omega_p}\ . \tag{9}$$

The Ginzburg-Landau κ near T_c is

$$\kappa=\frac{0.137(1+\lambda)^{3/2}T_c}{\Omega_p v_F x(z)} \tag{10}$$

and the temperature dependence of the upper critical field near T_c is given by

$$-\left.\frac{dH_{c2}}{dT}\right|_{T_c}=\frac{3.18(1+\lambda)^2T_c}{x(z)v_F}\ , \tag{11}$$

where

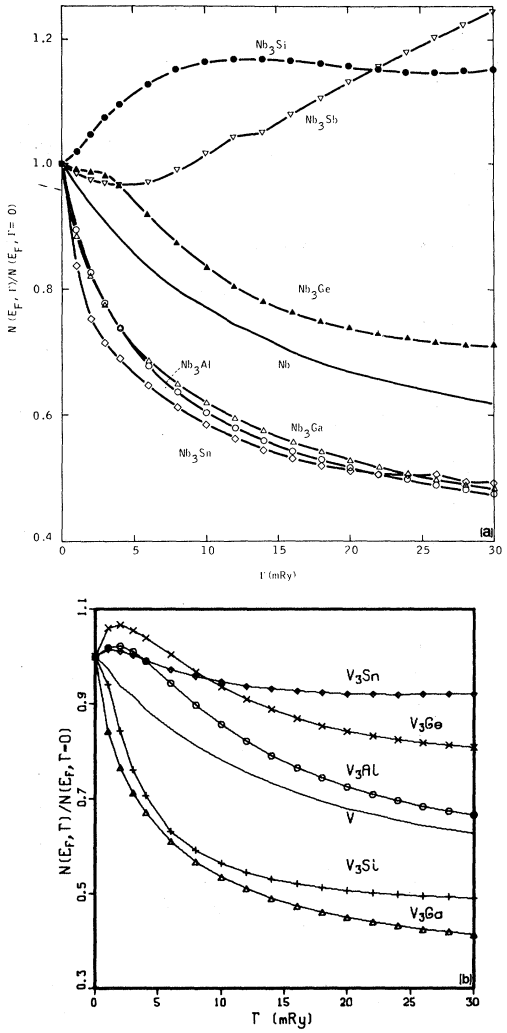

FIG. 1. Calculated density of states at the Fermi energy normalized to the density of states of the corresponding ordered material as a function of the electron linewidth Γ for (a) Nb and the Nb-based $A15$ compounds and for (b) V and the V-based $A15$ compounds.

$$z=\frac{0.88\xi_0}{l}=\frac{0.246\Omega_p^2\rho_0}{(1+\lambda)T_c}\ , \tag{12}$$

$$x(z)\approx\left[1+\frac{z}{1.173}\right]^{-1}\ , \tag{13}$$

where ξ_0,λ_L are in Å and dH_{c2}/dT is in Oe/K. Equations (10) and (11) do not include the strong-coupling corrections.[9]

In Table I we show the values of the different quantities for the perfectly ordered $A15$ compounds. T_{c0} is the experimental value and $\lambda^{\rm expt}$ is the experimentally measured λ found by inverting McMillan's equation. The rest of the parameters

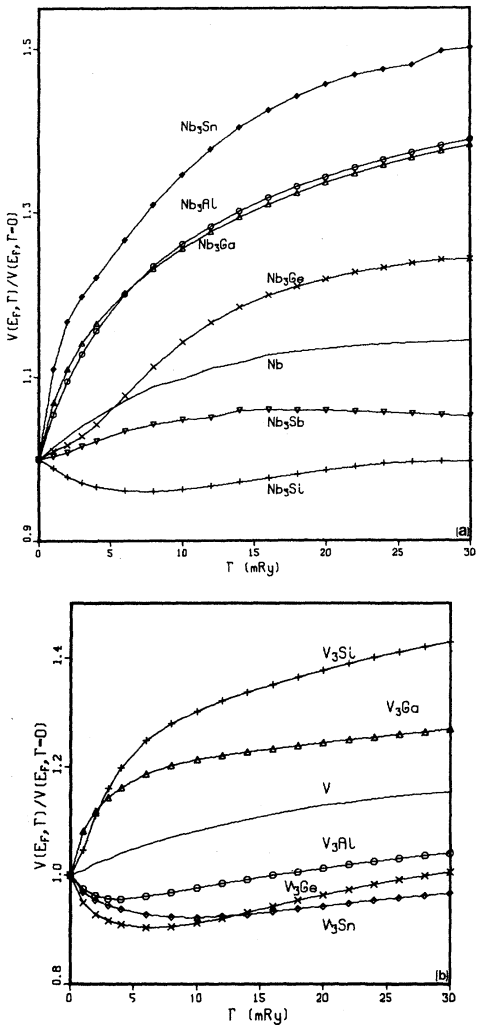

FIG. 2. Calculated Fermi velocities at the Fermi energy normalized to the Fermi velocity of the corresponding ordered material, as a function of the electron linewidth Γ for (a) Nb and Nb-based $A15$ compounds and for (b) V and V-based $A15$ compounds.

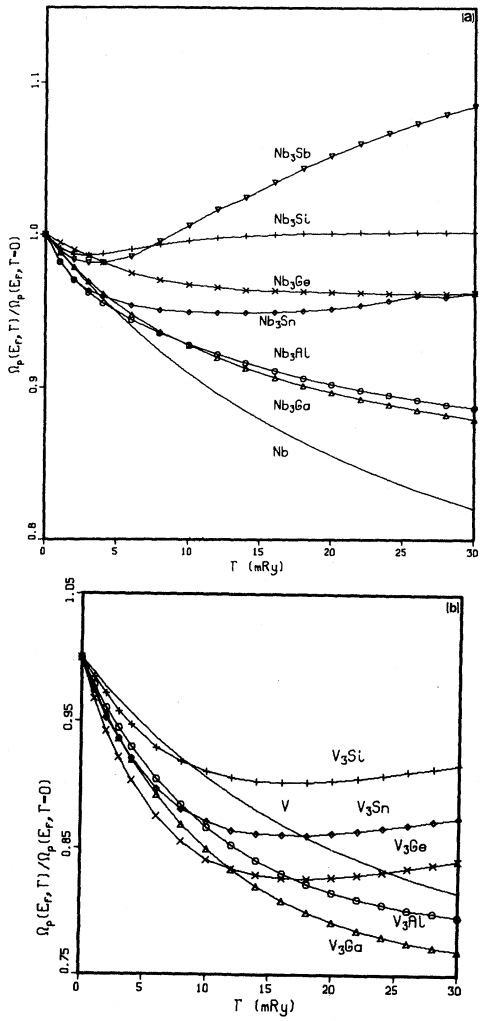

FIG. 3. Calculated plasma frequencies at the Fermi energy normalized to the plasma frequency of the corresponding ordered material as a function of the electron linewidth Γ for (a) Nb and the Nb-based $A15$ compounds and for (b) V and the V-based $A15$ compounds.

are those derived from band-structure calculations.[13]

Our results show that for all the $A15$ compounds we have studied except for Nb_3Sb and Nb_3Si, the DOS ultimately decreases as we increase the disorder. This seems to be expected for $A15$ compounds which are believed to exhibit a Fermi energy very near to a maximum of a sharp peak like V_3Si, Nb_3Al, Nb_3Sn, and V_3Ga. But we also find that $A15$ compounds exhibiting a Fermi energy *not* close to a peak in the DOS have a $N(E_F)$ which also decreases as we increase the disorder (V_3Al, V_3Ge, V_3Sn, and Nb_3Ge). Although in some cases, we have found a small increase of the $N(E_F)$ for low disorder further increase of Γ always reduced $N(E_F)$. For the low-T_c $A15$ Nb_3Sb we have a 20% increase in the DOS, while for Nb_3Si we also have a 15% increase. As we will discuss in more detail below, whatever was said above about $N(E_F)$ is applicable to the superconducting transition temperature T_c under the assumption that the electron-phonon coupling parameter λ is proportional to $N(E_F)$. This assumption seems to be reasonable and is supported by the calculations of Ref. 13. Using McMillan's equation one then gets T_c as a function of λ or ρ_0. While there seems to be a correlation between λ or T_c and $N(E_F)$ for the perfectly ordered materials, there is no such correlation be-

TABLE I. Measured superconducting temperature T_c^{expt}, experimentally derived value of the electron-phonon coupling λ^{expt}, and theoretical values at E_F of the Fermi velocity, plasmon energy, and density of states (Ref. 12).

	$T_{c0}=T_c^{\text{expt}}$ (K)	λ^{expt}	$v_F(E_F,\Gamma=0)$ (10^8 cm/sec)	$\Omega_p(E_F,\Gamma=0)$ (eV)	$N(E_F\Gamma=0)$ (per Ry per cell)
V	5.3	0.67	0.373	7.99	33.0
V_3Al	9.6	0.87	0.248	4.46	188.9
V_3Ga	16.5	1.17	0.205	4.63	295.6
V_3Si	17.1	1.12	0.210	4.02	200.2
V_3Ge	6.1	0.70	0.290	4.14	114.7
V_3Sn	3.8	0.61	0.270	3.73	123.0
Nb	9.2	0.94	0.565	9.45	26.3
Nb_3Al	18.6	1.60	0.277	4.58	199.2
Nb_3Ga	20.3	1.74	0.289	4.71	191.8
Nb_3Si	18.0	1.44	0.460	4.43	61.42
Nb_3Ge	23.2	1.80	0.356	4.35	106.7
Nb_3Sn	18.0	1.44	0.278	4.00	158.6
Nb_3Sb	0.2	0.38	0.451	3.74	51.95

tween T_c and the plasma frequency $\Omega_p(E_F)$ or the Fermi velocity v_F. There are relatively small variations of Ω_p and v_F among the $A15$ materials. The introduction of disorder, as described above, has little effect on $\Omega_p(E_F)$. There is always a decrease of the order of 10—20%, except for the case of Nb_3Sb where there is a 10% increase. These results appear to be reasonable since with increasing disorder less free carriers exist and so Ω_p decreases. Our results for Ω_p could be tested with optical data on pure and disordered $A15$ materials, but unfortunately, no such experimental results are available. Such optical measurements together with resistivity data would provide an independent test of the electron damping as a function of resistivity for disordered $A15$ systems. Finally v_F as a function of disorder Γ seems to increase as Γ increases for almost all the $A15$ compounds.

The superconducting transition temperature T_c was obtained by assuming that $\lambda \sim N(E_F,\Gamma)$ and using McMillan's equation with $\mu^*=0.13$ and Θ_D to be constant. We used

$$\lambda=\frac{\lambda^{\text{expt}}N(E_F,\Gamma)}{N(E_F,\Gamma=0)},$$

where λ^{expt} is the experimentally measured λ for the perfectly ordered material found by inverting McMillan's equation. Our results for T_c/T_{c0} vs ρ_0 are shown in Figs. 4(a) and 4(b) for the Nb- and V-based materials, respectively. For the Nb $A15$'s [Fig. 4(a)], we have found that T_c for Nb_3Ga, Nb_3Sn, and Nb_3Al decrease by 80% when ρ_0 reaches 150—200 $\mu\Omega$ cm in qualitative agreement with experiment.[2] Our detailed calculations for Nb_3Ge yield a smaller variation in T_c than observed experimentally. Ruvalds and Soukoulis[8] attributed the drop in T_c in Nb_3Ge to overdamping of acoustic plasmons. On this point Klein *et al.*[13] proposed an unusual phonon softening in Nb_3Ge in order to

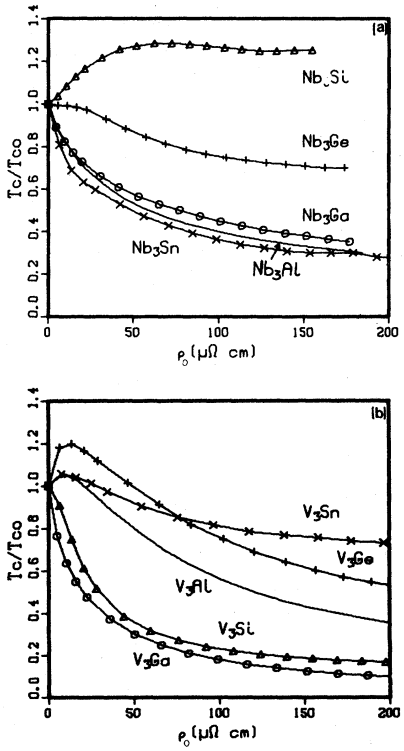

FIG. 4. Calculated transition temperatures normalized to the transition temperature of the corresponding ordered material as a function of the residual resistivity ρ_0.

reconcile the relatively low $N(E_F)$ with the high T_c. Recent tunneling experiments[15] strongly support this idea. Hence, for Nb_3Ge, the drop in T_c seen experimentally is not coming from the decrease of $N(E_F)$ but is probably a phonon effect. For Nb_3Si we predict that T_c will increase by 30% as disorder increases, which may explain the disagreement between experiment[16] and low-λ and -T_c values calculated at full stoichiometry.[13] For the V-based $A15$'s [Fig. 4(b)], we have found that the T_c drop in V_3Si and V_3Ga is in qualitative agreement with experiment.[3] The existing measured values of T_c for V_3Ge (Ref. 3) are also in qualitative agreement with our results, but due to difficulties in making clean samples there are no experimental values in the region with $\rho_0 \leq 50\ \hat{\mu}\Omega$ cm to check if the calculated initial increase in T_c with ρ_0 is real. For V_3Sn and V_3Al, the decrease of T_c is 20% and 60%, respectively. Insufficient or no data for the latter compounds prohibits us from comparing our results with experiments.

Our results for the temperature dependence of the upper critical field near T_c are shown in Figs. 5(a) and 5(b). For all the $A15$ compounds we have found that $|dH_{c2}/dT|$ increases as the disorder increases, which is the opposite of what the transition temperature T_c does with disorder. This trend was also seen in experiments done on Nb_3Al,[17] Nb_3Sn, V_3Si,[18] and V_3Ga.[19] We also want to mention that it is possible to get a higher upper critical field H_{c2} in the dirty limit ($\rho_0 > 0$) than the clean limit ($\rho_0 = 0$). This is so because the upper critical field is proportional to the product of the critical temperature T_c and dH_{c2}/dT as shown in the following equation:

$$H_{c2}(0) \approx T_c \left.\frac{dH_{c2}}{dT}\right|_{T_c} .$$

Therefore it is possible to get a higher H_{c2} for $\rho_0 > 0$ provided that dH_{c2}/dT increases faster with ρ_0 than T_c decreases with ρ_0. This argument is correct only in the absence of Pauli paramagnetic limiting process. Figures 6(a) and 6(b) show the mean free path l as a function of disorder. As expected, l for all the $A15$ compounds decreases with increasing disorder in a universal manner. In particular $l \approx 125-150$ Å for $\rho_0 \approx 10\ \mu\Omega$ cm and $l \approx 5-7$ Å for $\rho_0 \approx 150\ \mu\Omega$ cm. As one can see from Figs. 7(a) and 7(b) the $T=0$ BCS coherence length ξ_0 increases with disorder for almost all the $A15$ compounds. The only exception is Nb_3Sb, but its anomalous behavior is related to its very low T_c. By comparing Figs. 6 and 7 we see in the low disorder limit ($\rho_0 \approx 10\ \mu\Omega$ cm) none of the $A15$'s compounds satisfy the clean-limit approximation $l \gg \xi_0$. On the contrary $\xi_0 \approx l$. On the other hand in the high disorder limit our results satisfy the dirty-limit approximation that $\xi_0 \gg l$. Experimental results[16] for Nb_3Sn and V_3Si for l and ξ_0 show qualitatively the same behavior as our calculations. The Ginzburg-

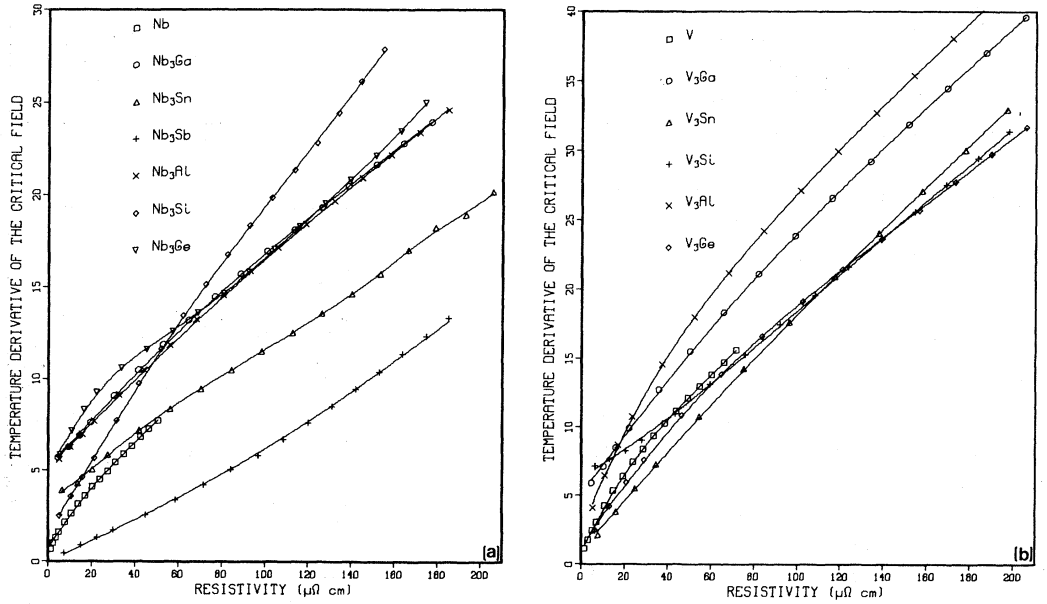

FIG. 5. Calculated temperature derivative of upper critical temperature near T_c in kOe/K as a function of the residual resistivity ρ_0.

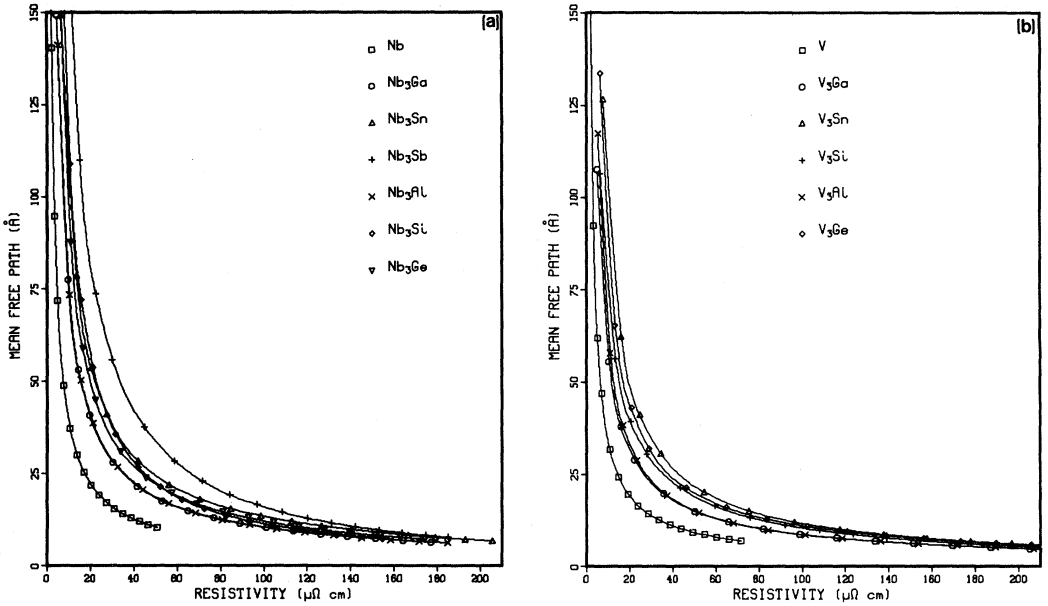

FIG. 6. Calculated mean free path in Å as a function of residual resistivity ρ_0.

Landau κ near T_c increases for all the $A15$'s with disorder as seen in Fig. 8. The increase in κ is almost an order of magnitude, provided that $\rho_0 \approx 180$ $\mu\Omega$ cm. In Fig. 9 we plot the $T=0$ London penetration depth λ_L versus disorder. Note that λ_L decreases or increases by a maximum of 15%, and this reflects the changes in the plasma frequencies.

Finally, we note that our results for V_3Si, Nb_3Ge, and Nb_3Sn are in at least qualitative agreement with those of Testardi and Mattheiss.[9] Small differences between the calculations are due to self-consistency effects and different treatment of exchange in the band-structure calculations; and also due to the fact that we have used a Lorentzian broadening rather than the thermal broadening used by Testardi and Mattheiss.

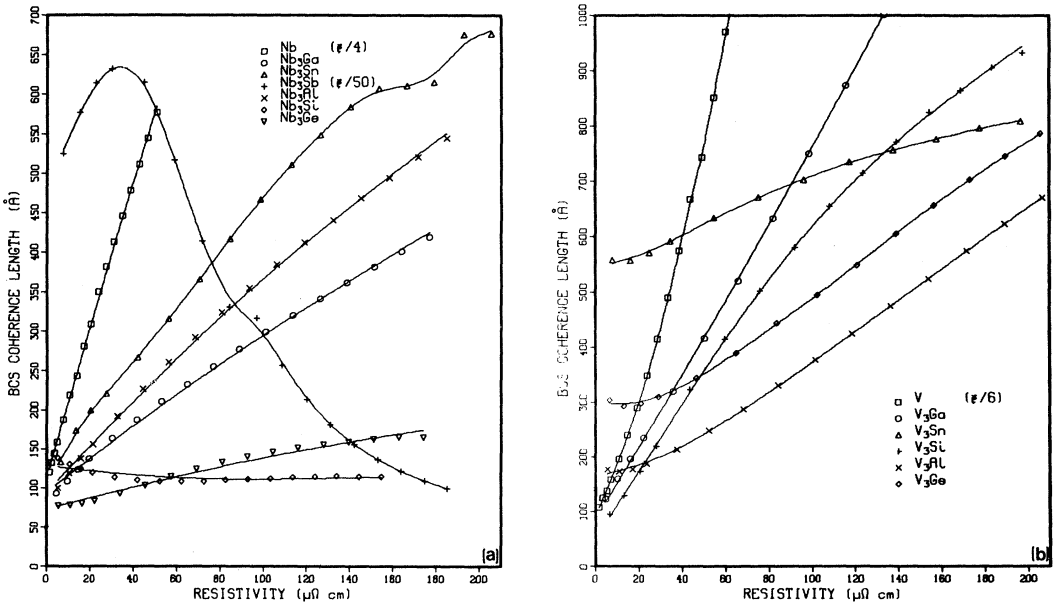

FIG. 7. Calculated BCS coherence length at 0 K in Å as a function of resistivity ρ_0.

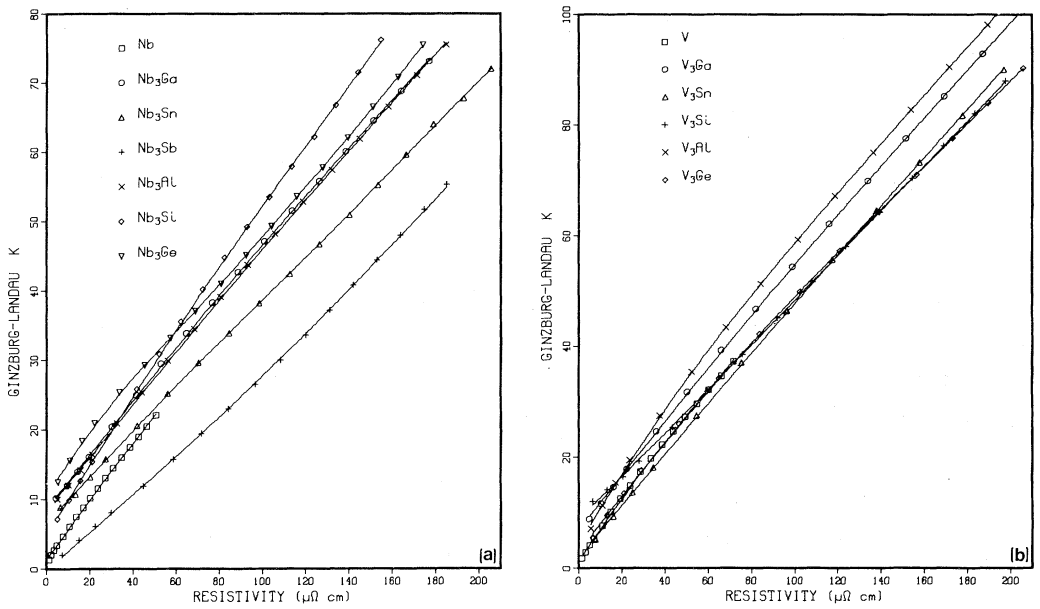

FIG 8. Ginzburg-Landau κ near T_c as a function of residual resistivity ρ_0.

IV. CONCLUSIONS

To summarize our findings, using the electronic band-structure calculations for the DOS, Fermi velocity, and plasma frequency for a series of $A15$ materials, we have calculated the $N(E_F)$, $\Omega_p(E_F)$, v_F, dH_{c2}/dT, l, ξ_0, κ, and λ_L as a function of disorder. Disorder is introduced by averaging the above equations over an energy internal $\Gamma=\hbar/\tau$ around the Fermi level. We found that for the $A15$ materials studied (except for Nb_3Sb and Nb_3Si), $N(E_F)$ decreases as we increase disorder. By assuming that $\lambda \sim N(E_F)$, and using McMillan's equation for T_c we find that T_c always decreases (except for Nb_3Sb and Nb_3Si) with the introduction of disorder. Our detailed calculations suggest that the drop of T_c in

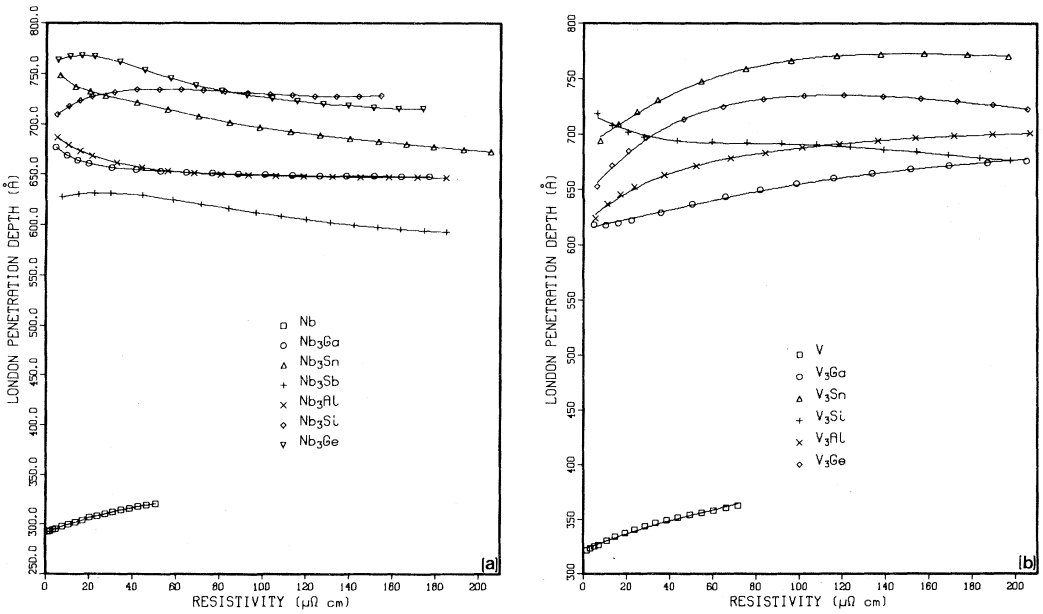

FIG. 9. Calculated London penetration depth at 0 K in Å as a function of residual resistivity ρ_0.

the $A15$ superconductors is due to changes of the DOS, in particular the decrease of $N(E_F)$ with disorder. Magnetic susceptibility and specific-heat data on pure and disordered $A15$ compounds (especially for Nb_3Ge and V_3Al which do not have their E_F's at a peak) will be useful for finding out changes in the DOS and whether or not a temperature-dependent susceptibility exists. The behavior of T_c vs ρ_0 is in qualitative agreement with most of the existing experiments for $N(E_F)$ and T_c. Optical data on pure and disordered $A15$ materials in connection with resistivity data will give an independent estimate of the electron width Γ as a function of resistivity.

We now wish to comment on the prospects of achieving higher superconductivity temperatures by shifting the Fermi energy to a favorable maximum in $N(E_F)$. This could be accomplished by alloying the $A15$ compounds and thus create $A15$ pseudobinaries.[13] However, in this situation $N(E_F)$ may not reach a high value predicted by the rigid-band model but instead alloying can cause a reduction of the electron lifetime, and thus a reduction of $N(E_F)$. Another possibility may be to change the electronic structure by ion implantation of hydrogen in compounds like Nb_3Ge, Nb_3Si, and V_3Sn, with the goal of increasing $N(E_F)$, provided that the induced disorder is below a prescribed limit. However, the explorations for achieving a higher $N(E_F)$ need to be coupled with the phonon behavior, because it is possible that mechanisms which may enhance $N(E_F)$ could harden the phonon modes and therefore prevent the raising of T_c. Finally, it would also be interesting to calculate $N(E)$ for the disordered $A15$ compounds using the coherent-potential approximation and thus remove the uncertainties of the electron-lifetime model.

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