Microscopic superconducting parameters of Nb_3Al: Importance of the band density of states

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Critical-field measurements are carried out in low fields on a series of $A15$ Nb-Al as-deposited films where structural disorder, as characterized by the residual resistivity $\rho(T_c)$, is introduced by varying the composition in the $A15$ phase. Analysis of the critical-field slopes near T_c gives relatively low electronic densities of states at Fermi level $N^b(0)$ in agreement with heat-capacity results. The $N^b(0)$'s remain nearly constant over the resistivity range varied. Lacking data in the low resistivity range, our results neither support nor disagree with the resistive lifetime-broadening model of the density of states. Nonetheless, the data unambiguously demonstrate that the conventional model, in which a peak in the band density of states at the Fermi level plays the essential role, is not sufficient to explain the observed systematics in high-T_c Nb_3Al. More specifically, changes in $\langle I^2\rangle/\langle\omega^2\rangle$ with disorder are found to be responsible for the observed changes in T_c, where the increase of $\langle\omega^2\rangle$ with disorder has been shown earlier in the tunneling studies. Further consideration of similar data for the other superconductors V_3Si, Nb_3Sn, and Nb_3Ge suggests that important variations in $\langle I^2\rangle/\langle\omega^2\rangle$ may be a general feature of the high-T_c $A15$ superconductors. $\langle I^2\rangle/\langle\omega^2\rangle$ increases as one progresses from V_3Si through Nb_3Sn and Nb_3Al to Nb_3Ge in correlation with the relative instability of the $A15$ phase in these materials.

I. INTRODUCTION

The $A15$ superconductors have received considerable interest because of the favored occurrence of high-critical-temperature superconductivity in this class of compounds. However the fundamental properties with which the high T_c's of these materials can be associated remain unsettled. In the conventional model[1] based on the properties of the well-ordered, stoichiometric compounds such as Nb_3Sn and V_3Si, a sharp peak in the band density of states is believed to play an important role both in the high-T_c superconductivity of these materials and also in their anomalous normal-state properties. However, the so-called metastable $A15$ compounds (i.e., those that do not exist in thermodynamic equilibrium at stoichiometry) such as Nb_3Al and Nb_3Ge of even higher T_c do not show such anomalous normal-state behavior in the heat capacity[2,3] and magnetic susceptibility[4] and therefore are in potential conflict with the conventional model. On the other hand they have not been made as well-ordered, stoichiometric compounds, and clear evidence for inhomogeneities in these materials has been observed,[5] further complicating any interpretation. Thus a definite conclusion about the origin of the high T_c's in these superconductors cannot yet be reached.

An alternative in comparing the properties among different compounds is introducing disorder or defects artificially[6] and even including the amorphous counterpart[7] in a systematic study of a given compound. The trends in the physical properties provide additional insights into the mechanisms responsible for the high-T_c superconductivity. In the case of the canonical $A15$ superconductor V_3Si, the depression of T_c with increasing disorder has been correlated with a decreasing band density of states at the Fermi level $N^b(0)$.[8,9] For metastable Nb_3Ge, a large reduction in the ratio $\lambda/N^b(0)$ or $\langle I^2\rangle$ with disorder has been suggested by Tsuei,[7] and Ghosh and Strongin.[9] Here λ is the electron-phonon interaction parameter and $\langle I^2\rangle$ is the average squared electron-phonon matrix element. However these conclusions become less certain when one recognizes the unknown or uncontrollable degree of inhomogeneities present in the metastable materials. Also (taken by themselves) the interpretations from such studies are subject to uncertainities resulting from the assumptions made about the lattice properties in the analysis used to obtain the microscopic parameters.

Given the above difficulties, we have undertaken a comprehensive study of the least unstable member of the metastable class of $A15$ high-temperature superconductors, namely, Nb_3Al, as a promising prospect for improving understanding. The synthesis of this compound in thin film form such as used in this study is well-controlled as described in a previous publication.[10] Homogeneous samples with systematic and reproducible material characteristics across the single-phase $A15$ phase field (i.e., from 19 to 22.6

at.% Al at low temperatures) can be prepared. Superconducting tunneling measurements[11] carried out on $A15$ Nb-Al thin films as a function of Al composition establish for the first time the important role of soft phonons in the high-T_c superconductivity. However the basic mechanisms of the superconductivity cannot be fully understood without additional information regarding the electronic properties. Therefore in the present work we extend our study of the Nb_3Al system to include critical-field measurements on a series of Nb-Al films varying in composition across the single-phase field. Along with our tunneling measurements we can determine the various microscopic parameters [e.g., $N^b(0)$, $\langle I^2\rangle$, and $\langle\omega^2\rangle$, etc.] for understanding superconductivity.

Our analysis yields a relatively low value of $N^b(0)$ for Nb_3Al, in agreement with the heat-capacity results.[2,12] Moreover, strikingly, the value of $N^b(0)$ remains approximately constant over the compositional range varied. On the other hand, a substantial decrease of $\langle I^2\rangle$ and increase of $\langle\omega^2\rangle$ (i.e., stiffening of the phonon modes) account for the depression of T_c with disorder. The possibility of a variation of $\langle I^2\rangle/\langle\omega^2\rangle$ in Nb_3Al with disorder has been noted earlier based on a similar study on radiation-damaged Nb_3Al by Ghosh and Strongin.[9] This conclusion has not been universally accepted, however. The present work based on a very thorough analysis and particularly well-characterized samples (including electron tunneling) now leaves little doubt that variation in $\langle I^2\rangle$ with disorder is an essential feature of the $A15$ Nb_3Al. Comparing these results to parallel studies on other $A15$ superconductors shows a systematic progression of the ratio $\langle I^2\rangle/\langle\omega^2\rangle$ among V_3Si, Nb_3Sn, Nb_3Al, and Nb_3Ge that correlates with the relative $A15$ phase instability of these compounds.

II. SAMPLE PREPARATION AND EXPERIMENTAL PROCEDURES

The Nb_3Al thin films used in this study were prepared by the technique of electron-beam coevaporation at the optimum deposition conditions as determined in the earlier synthesis work.[10,13] Such optimized deposition conditions give Nb-Al films with lattice parameters following the prediction of Geller radii, suggesting that the $A15$ structure produced is as ordered as allowed by the composition. Hence the variations of the T_c's and residual resistivities $\rho(T_c)$'s (which we take as an approximate measure of the structure disorder), of the samples investigated in this work are caused mainly by variations in the Al composition.

In order to carry out the four-point resistance measurements used to determine the resistivity and the critical field, the films were etched into a suitable bridge pattern using photolithography. Since the chemical etch used in the patterning contained hydrofluoric acid, which has been shown to be detrimental to the metastable $A15$'s,[14] the T_c's were compared for one sample before and after a 5-sec etch and showed no apparent degradation. The thicknesses were measured by a mechanical stylus technique and are in the range from 5000 to 6000 Å.

All the critical fields were measured with the field both parallel and perpendicular to the film. The critical fields were determined from both dc and ac resistive transitions obtained while sweeping the temperature in a fixed field. Since the transitions of most samples tended to broaden in high fields (e.g., from 0.4 to 2.0 K as the field increased up to 80 kOe), the transition temperature was chosen to be the midpoint of the resistive transition throughout all measurements.

III. RESULTS AND ANALYSIS

A. Critical fields and transition temperatures

Representative critical-field data of our samples at low fields near T_c are shown in Fig. 1. The low-field data of most samples show a rather linear dependence of H_{c2} on temperature. A slight curvature exists in the data very near T_c, deviating from the straight lines expected from Ginzburg-Landau-Abrikosov-Gorkov (GLAG) theory.[15] The difference between the measured and extrapolated T_c's are at most 0.1 K, however, despite these small anomalies, the critical-field slope $(dH_2/dT)_{T_c}$ [from which $N^b(0)$ is derived] could be determined satisfactorily from the linear portion of the data between 20 and 80 kOe. The $(dH_{c2}/dT)_{T_c}$ for all the samples studied along

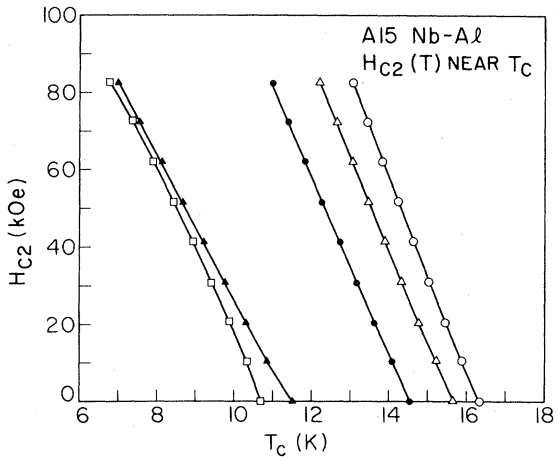

FIG. 1. Representative critical-field data near T_c of the $A15$ Nb-Al films measured. The lines drawn through data points are intended to serve only as a guide to the eye.

with other pertinent material parameters are listed in Table I.

The dependence of T_c and $(dH_{c2}/dT)_{T_c}$ on residual resistivity $\rho(T_c)$ for our Nb-Al samples are shown in Figs. 2(a) and 2(b) along with similar data for radiation damaged[9] Nb_3Al. Also shown for comparison are equivalent sets of data for the Nb_3Sn system.[6,9,16]

Our Nb-Al data in general show a trend of decreasing T_c and $(dH_{c2}/dT)_{T_c}$ with increasing resistivity. The data on a bulk Nb_3Al sample,[17] whose resistivity is determined by normalizing its resistivity ratio to the same room-temperature resistivity ($81 \pm 2\ \mu\Omega$ cm), agrees with the linear trend seen in our thin-film results for T_c vs $\rho(T_c)$. The earlier critical-field measurement[18] on a bulk Nb_3Al sample with $T_c = 18.7$ K gives a $(dH_{c2}/dT)_{T_c}$ of 25.6 kOe/K comparable to our results as well.

The data for radiation-damaged[9] Nb_3Al show a less rapid degradation of T_c with $\rho(T_c)$ than our data on Nb-Al in which composition was varied, although the

TABLE I. Material parameters of the Nb-Al samples. Samples are identified individually to allow specific comparison with other physical measurements carried out or to be carried out on these samples and reported elsewhere.

Sample No.	$T_c(H_{c2})$	(at.% Al)	$\rho(T_c)$ ($\mu\Omega$ cm)	$\left(\frac{dH_{c2}}{dT}\right)_{T_c}$ (kOe/K)	$\eta_{H_{c2}}(T_c)$	λ
G-78-18 No. 8C-I	16.24	22.7	54.52	25.66	1.19	1.65
G-80-41 No. 4B-I	16.03	22.6	57.78	26.41	1.18	1.60
G-80-41 No. 4B-II	15.97	22.5	56.37	26.78	1.18	1.60
G-78-18 No. 7C-I	15.60	22.2	54.66	24.41	1.144	1.54
G-78-18 No. 7C-II	14.93	21.9	55.27	23.13	1.122	1.42
G-77-59 No. 5A-I	14.52	21.7	63.13	23.07	1.071	1.36
G-77-59 No. 5A-II	14.47	21.7	60.97	22.91	1.071	1.36
G-78-18 No. 5C-I	11.44	20.1	62.70	18.56	1.0	1.12
G-78-18 No. 5C-II	11.01	19.9	63.16	19.47	1.0	1.10

Sample No.	S/S_F	γ (mJ/cm^{3}K^2)	$N^*(0)$ $\left(\frac{\text{states}}{\text{eV spin unit cell}}\right)$	$N^b(0)$ $\left(\frac{\text{states}}{\text{eV spin unit cell}}\right)$
G-78-18 No. 8C-I	0.37±0.15	$0.71 \pm^{0.06}_{0.12}$	$12.66 \pm^{1.05}_{2.13}$	$4.78 \pm^{0.40}_{0.81}$
G-80-41 No. 4B-I	0.37±0.15	$0.71 \pm^{0.06}_{0.12}$	$12.55 \pm^{0.98}_{2.13}$	$4.83 \pm^{0.38}_{0.82}$
G-80-41 No. 4B-II	0.37±0.15	$0.73 \pm^{0.06}_{0.12}$	$12.89 \pm^{1.05}_{2.13}$	$4.96 \pm^{0.40}_{0.82}$
G-78-18 No. 7C-I	0.37±0.15	$0.71 \pm^{0.06}_{0.12}$	$12.60 \pm^{1.00}_{2.13}$	$4.96 \pm^{0.39}_{0.84}$
G-78-18 No. 7C-II	0.37±0.15	$0.68 \pm^{0.05}_{0.11}$	$12.12 \pm^{0.94}_{1.96}$	$5.01 \pm^{0.39}_{0.81}$
G-77-59 No. 5A-I	0.37±0.15	$0.64 \pm^{0.04}_{0.09}$	$11.30 \pm^{0.73}_{1.60}$	$4.79 \pm^{0.31}_{0.68}$
G-77-59 No. 5A-II	0.37±0.15	$0.65 \pm^{0.04}_{0.10}$	$11.56 \pm^{0.78}_{1.78}$	$4.90 \pm^{0.33}_{0.76}$
G-78-18 No. 5C-I	0.37±0.15	$0.58 \pm^{0.03}_{0.07}$	$10.35 \pm^{0.52}_{1.25}$	$4.88 \pm^{0.25}_{0.60}$
G-78-18 No. 5C-II	0.37±0.15	$0.61 \pm^{0.30}_{0.07}$	$10.77 \pm^{0.55}_{1.30}$	$5.13 \pm^{0.26}_{0.62}$

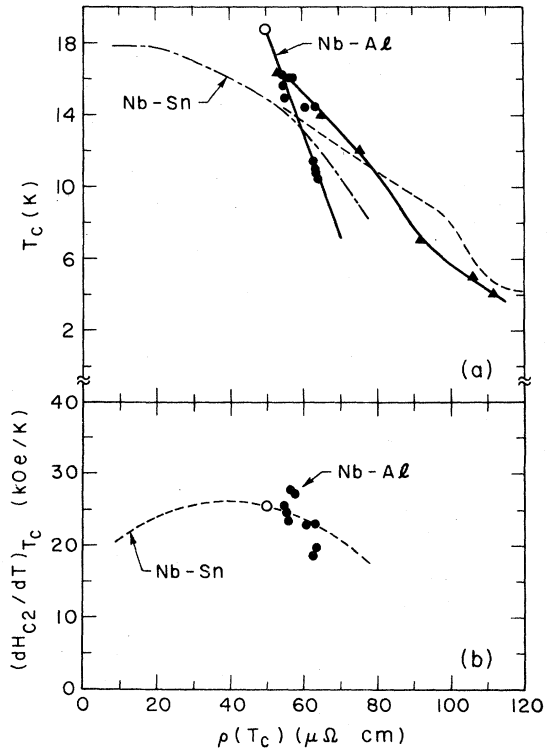

FIG. 2. (a) T_c vs $\rho(T_c)$ and (b) $(dH_{c2}/dT)_{T_c}$ vs $\rho(T_c)$ for the Nb_3Al and the Nb_3Sn systems: (○) bulk Nb_3Al, • (●) Nb-Al this work, (▲) Nb_3Al radiation damaged, (– – –) Nb-Sn (Ref. 16), and (–●–) Nb_3Sn radiation damaged (Refs. 6 and 9).

initial material before damage in the work of Ref. 9 was prepared in a similar fashion as in this work and has comparable T_c, $\rho(T_c)$, and $(dH_{c2}/dT)_{T_c}$. A similar deviation in the T_c vs $\rho(T_c)$ data between the cases of radiation damage[6,9] and of varying composition[16] is seen for Nb_3Sn. In fact it occurs at comparable resistivities ($\gtrsim 55$ $\mu\Omega$ cm) and at the comparable compositions (≤ 23 at.% of Sn or Al). The absence of universal behavior in the depression of T_c with structural disorder for $\rho(T_c) \gtrsim 55$ $\mu\Omega$ cm, which is the range over which most Nb_3Al samples actually exist, tends not to support the existence of a definitive "universal defect"[19] as the source of the depression of T_c.

Compared to the Nb_3Sn system, the data on Nb_3Al show a much more sensitive dependence of T_c on $\rho(T_c)$ independently of whether the disorder is introduced by applying radiation damage or by varying the $A15$ composition. In addition, the $(dH_{c2}/dT)_{T_c}$ data of Nb-Sn exhibit a maximum value near a $\rho(T_c)$ of about 40–50 $\mu\Omega$ cm which is not evident for Nb-Al. Note that $\rho(300\text{ K}) \simeq 90$ $\mu\Omega$ cm of Nb_3Sn is slightly larger than that of Nb_3Al. Extrapolating the Nb-Al data back to $\rho(T_c) \sim 0$ as guided by the features of the Nb-Sn data suggests a $T_c \gtrsim 21$ K for very clean Nb_3Al if it could be made.

The existence of a very high T_c and its sensitive degradation with disorder has been attributed[6,8,9] for the $A15$ V_3Si and Nb_3Sn to the sharp feature in the band density of states near the Fermi level. Whether such a model accounts for the even more rapid depression of T_c with $\rho(T_c)$ in the Nb_3Al requires a detailed examination of the microscopic parameters by extracting the $N^b(0)$'s from the measured $(dH_{c2}/dT)_{T_c}$'s. The procedures and results are described in the next section.

B. Densities of states at the Fermi Level $N^b(0)$

Based on the data of the critical-field slope near T_c, the general procedure of evaluating various superconducting and normal-state parameters including $N^b(0)$ is well formulated.[20] Briefly, the slope of critical field near T_c including corrections for the electron-phonon interaction can be written as[21]

$$\left.\frac{dH_{c2}}{dT}\right|_{T_c} = \eta_{H_{c2}}(T_c)\left[9.55\times10^{24}\gamma^{*2}T_c\left(\frac{n^{3/2}S}{S_F}\right)^{-2} + 5.26\times10^{4}\gamma^{*}\rho(\Omega\text{ cm})\right]\times[R(\gamma_{tr})]^{-1}\text{ Oe/K}\ , \quad (1)$$

where $\eta_{H_{c2}}(T_c)$ is a strong-coupling factor for the magnetic pair-breaking parameter,[22,23] of order of $1.0 \sim 1.2$; γ^* is the renormalized electronic heat-capacity coefficient in erg/cm^{3}K^2 and S/S_F is the ratio of the Fermi surface S to the corresponding Fermi surface S_F of a free-electron gas of density n. Once $(dH_{c2}/dT)_{T_c}$, T_c, $\rho(T_c)$ are measured together with suitable estimates of $\eta_{H_{c2}}(T_c)$ and S/S_F, γ^* can then be obtained.[24]

To estimate $\eta_{H_{c2}}(T_c)$ information about the strong-coupling properties of the material is required. This is available from the tunneling studies carried out previously on similar Nb_3Al thin films.[11] The coupling strength $2\Delta/k_BT_c$ varies linearly from approximately the BCS value to 4.4 as the composition varies from 21.5 to 22.8 at.% Al and T_c increases from 14.0 to 16.4 K. Thus $\eta_{H_{c2}}(T_c)$ must be determined for each individual sample with different T_c and composition. This is done by using the mea-

sured $2\Delta/k_BT_c$ to calculate (T_c/ω_0) from the analytical expression of Kresin and Parkhomenko[23] based on a simple Einstein spectrum for $\alpha^2F(\omega)$ and then using this value T_c/ω_0 to calculate the $\eta_{H_{c2}}(T_c)$. For samples with T_c less than 14.0 K where $2\Delta/k_BT_c$ is BCS-like, $\eta_{H_{c2}}(T_c)$ is taken to be 1.0.

The S/S_F is determined by requiring the extrapolated γ^{*M} from the results derived magnetically in this work to agree with the thermally derived γ^{*c} corresponding to a T_c of 18.7 K. In this procedure we have assigned a $\rho(T_c)$ of 50 $\mu\Omega$ cm for the sample in that heat-capacity measurement according to the systematic trend T_c vs $\rho(T_c)$ shown in Fig. 2(a). The S/S_F ratio is thus found to be 0.37. In the dirty limit, which is the case for all Nb_3Al samples measured, the second term in Eq. (1) dominates and the uncertainty in the factor S/S_F causes only a small uncertainty in the determination of γ^{*M}'s.

The resultant γ^{*M}'s from this analysis are shown in Fig. 3(a) as a function of the residual resistivity $\rho(T_c)$ and are listed in Table I. The asymmetric "error bars" in the γ^{*M}'s represent the uncertainty due to S/S_F of 0.37 $\pm$0.15. A further increase of $S/S_F > 0.52$ only slightly increases γ^{*M}. Therefore, within the accuracy of our estimates, a large γ^* comparable to those of the stoichiometric and ordered Nb_3Sn and V_3Si is unlikely in the presently best-made Nb_3Al even with a T_c onset $\sim$19.0 K. The dashed line shows γ^{*M} vs $\rho(T_c)$ for Nb-Sn samples[16] analyzed by the similar method which includes a possible increase of $\langle\omega\rangle$ with disorder. The reason for doing so will be discussed in detail in a later section.

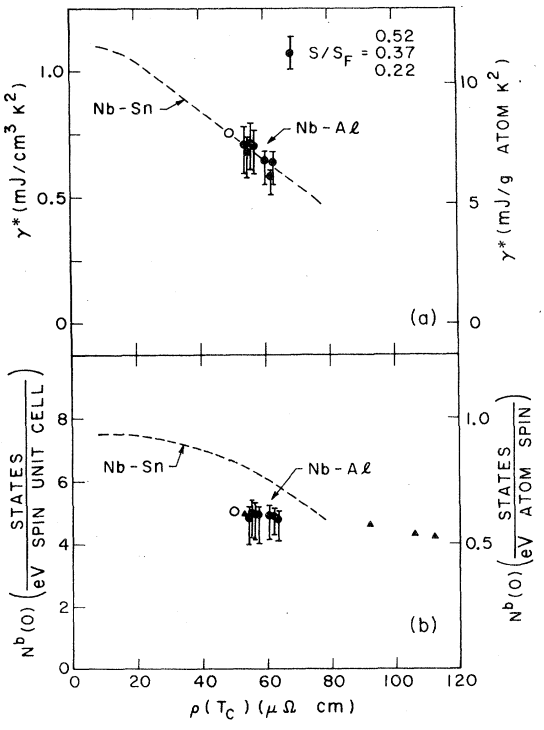

FIG. 3. (a) Electronic specific-heat coefficient γ^* vs $\rho(T_c)$ and (b) band density of states at the Fermi level $N^b(0)$ vs $\rho(T_c)$, for (○) bulk Nb_3Al, (●) Nb-Al this work, (– – –) Nb-Sn Ref. 16, and (▲) Nb_3Al radiation damaged with corrections described in the text. The right-hand scale of γ^* in mJ/g atom K^2 is for Nb-Al only.

The band density of states for $A15$ Nb-Al can be calculated from γ^{*M}'s with the relation

$$N^b(0)\left(\frac{\text{states}}{\text{eV spin unit cell}}\right)=\frac{17.8}{1+\lambda}\gamma^{*M}\left(\frac{\text{mJ}}{\text{cm}^3\text{K}^2}\right), \quad (2)$$

where we take 5.180 Å to be the lattice parameter of $A15$ Nb_3Al. To estimate λ, we have used our tunneling data rather than Kresin and Parkhomenko's formulas,[23] since in the latter method the resolution in determining T_c/ω_0 in the weak-coupled limit is severely reduced.[25] From the gap inversion analysis in the tunneling studies,[11] $\langle\omega_{\log}\rangle$ and λ have been generated for two junctions with a T_c of 14.0 and 16.4 K. As shown by Allen and Dynes,[26] a nearly linear dependence of $T_c/\langle\omega_{\log}\rangle$ on λ over a small range of λ is found true for three possible spectra of $\alpha^2F(\omega)$. Hence λ can be obtained for samples of T_c between 14.0 and 16.4 K by interpolation for each $T_c/\langle\omega_{\log}\rangle$, where a linear decrease of $\langle\omega_{\log}\rangle$ is assumed to account for the linear increase in $2\Delta/k_BT_c$. For samples of $T_c < 14.0$ K, $\langle\omega_{\log}\rangle$ is taken to be the same as that of $T_c = 14.0$ K because the coupling strength remains BCS-like.

The resultant $N^b(0)$'s for the $A15$ Nb-Al films are plotted in Fig. 3(b) as denoted by the solid circles. As listed explicitly in Table I, the $N^b(0)$'s remain essentially constant over the T_c and $\rho(T_c)$ range investigated. The changes of $N^b(0)$ with $\rho(T_c)$ for the Nb-Sn samples[16] analyzed by the similar fashion are also shown in Fig. 3(b) by the dashed line for comparison. Note that it is the more rapid variation of the electron-phonon correction $(1+\lambda)$ with composition in Nb_3Al than Nb_3Sn over the range studied that has practically eliminated the variation in γ^* vs $\rho(T_c)$ seen in Fig. 3(a). The $N^b(0)$ data of the α-particle-damaged[9] Nb_3Al agree well with those of our Nb-Al samples of comparable T_c's, if one includes the corrections for the $\langle\omega\rangle$'s, λ's, and $\eta_{H_{c2}}(T_c)$'s as described earlier. In particular we take $\eta_{H_{c2}}(T_c)$ to be 1.0 for the weak-coupled samples with $T_c \leq 14.0$ K. The $N^b(0)$ vs $\rho(T_c)$ data after these corrections are shown in Fig. 3(b) by the solid triangles. Thus, even with a much larger variation of resistivity $\rho(T_c)$ or disorder up to 112 $\mu\Omega$ cm, $N^b(0)$ still remains nearly constant and only decreases by 15% at the extreme level of damage [$\rho(T_c) \sim 112$ $\mu\Omega$ cm].

IV. DISCUSSION

A. Microscopic parameters of Nb_3Al

The degradation of superconductivity due to disorder or defects in the high-T_c $A15$ superconductors has been intensively studied both theoretically[27] and experimentally.[6,9,19] The prevailing model[6,28] explaining such behavior is based on the idea that resistive-lifetime broadening of the density of states with the structural disorder is the fundamental factor in reducing T_c. However, in the case of the $A15$ Nb_3Al, a rapid degradation of T_c with disorder is found also (nearly twice as fast as those of Nb_3Sn) in both our Nb-Al films and the radiation damaged[9] Nb_3Al, and yet the $N^b(0)$'s derived from the analysis of the critical-field data remain approximately constant. Thus changes in $N^b(0)$ cannot account for the depression of T_c from 16.2 to 4 K. Hence in the McMillan equation $\lambda = N(0)\langle I^2\rangle / M\langle\omega^2\rangle$, λ cannot be simply proportional to $N^b(0)$, and a substantial decrease in the ratio $\lambda/N^b(0) \propto \langle I^2\rangle/\langle\omega^2\rangle$ (by as much as 50%) must be involved. As shown explicitly in the tunneling studies, the decrease of T_c from 16.4 to 14.0 K with the reduction of the ratio $2\Delta/k_BT_c$ from 4.4 to a BCS-like value mainly results from the phonon mode stiffening by 25%. If we assume $\langle\omega^2\rangle$ remains constant for $T_c \leq 14.0$ K where the coupling strength stays weakly coupled, the reduction in the ratio $\langle I^2\rangle/\langle\omega^2\rangle$ mostly arises from a decrease of $\langle I^2\rangle$ by 40%. These variations of the microscopic superconducting parameters, λ, $N^b(0)$, $\lambda/N^b(0)$, $\langle\omega^2\rangle$, and $\langle I^2\rangle$ for Nb_3Al with respect to T_c are shown in Fig. 4(a) by the solid line. Corrections related to any possible continuing increase of $\langle\omega^2\rangle$ with disorder for $T_c < 14.0$ K affects only the relative weightings between $\langle I^2\rangle$ and $\langle\omega^2\rangle$.

The modest value of $N^b(0)$ in $A15$ Nb_3Al and its insensitivity to structural disorder raises the question of the relative importance of $N^b(0)$ to the high-T_c superconductivity of Nb_3Al. As a consequence it is reasonable to ascertain how sensitive such results are to the analysis used, particularly since we include the unconventional feature of changes in $\langle\omega\rangle$'s with disorder in the analysis. In Fig. 4(a), we show the resultant microscopic parameters (dotted curves) analyzed assuming no phonon softening following Orlando *et al.*[21] The data for α-particle-damaged Nb_3Al analyzed by Ghosh and Strongin[9] (dash-dotted curves) are also shown. These latter authors used a larger $\langle\omega_{\log}\rangle$ to estimate λ, as suggested from the neutron work,[29] and took $\langle\omega_{\log}\rangle$ to be constant throughout. They reached the similar conclusion of the decrease of $\langle I^2\rangle/\langle\omega^2\rangle$ with disorder by about 25%. Comparing these three different analyses, the following conclusions can be drawn quite unambiguously: Any variation in $N^b(0)$ with T_c is indeed rather mild and can even be considered negligible. As a result a substantial decrease of $\langle I^2\rangle$ from 25% to as much as 55% is necessary to account for the lowering of T_c.

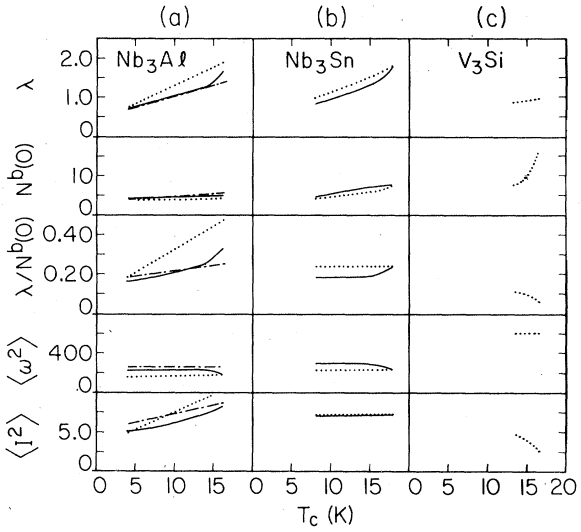

FIG. 4. Microscopic parameters λ, $N^b(0)$, $\lambda/N^b(0)$, $\langle\omega^2\rangle$, and $\langle I^2\rangle$ as a function of T_c for various $A15$ compounds where $N^b(0)$, $\langle\omega^2\rangle$, and $\langle I^2\rangle$ are in units of states/eV spin unit cell, $(\text{meV})^2$, and $(\text{eV/Å})^2$, respectively. The solid lines shown are the data analyzed by the method in this paper. The dotted lines are for the same data analyzed by assuming a constant $\langle\omega\rangle$ as in Ref. 21. The dash-dotted line corresponds to the data of radiation damaged Nb_3Al analyzed by Ghosh and Strongin (Ref. 9).

The controversy regarding the heat-capacity results[2] of $A15$ Nb_3Al can be reconciled based upon the above conclusions, since we find $N^b(0)$ remains nearly constant regardless of the amounts or the kinds of defects introduced. The increase in γ^* for Nb_3Al of even improved quality, as gauged by the narrower transition width in the heat-capacity measurement, is expected not to be substantial. This has been demonstrated in the more recent heat-capacity measurements[12,30] on Nb_3Al with a T_c onset 18.7 or 19.0 K and a relatively narrow transition width of $\sim$0.50 K. Furthermore, the heat-capacity measurement on the radiation damaged Nb_3Al ($T_c = 7.8$ K) of Cort *et al.*[12] showed a slight decrease of $N^b(0)$ and a substantial increase of the Debye temperature, thus supporting the stiffening of phonons with disorder that we found in the tunneling studies.

B. Comparison with other $A15$ superconductors

To understand further the systematics in the microscopic parameters of Nb_3Al, we compare our results with other high-T_c $A15$ compounds. The systematic changes in the parameters of Nb_3Sn obtained

by carrying out similar analyses on critical-field data near T_c are shown in Fig. 4(b).[16,21] The analysis[21] which assumes a constant $\langle\omega\rangle$ leads to an essentially constant ratio $\lambda/N(0)$ and hence a constant $\langle I^2\rangle$ over a variation of T_c from 17.8 to 8 K. However, tunneling studies in both Nb_3Sn (Ref. 31) and Nb_3Al (Ref. 11) find that the coupling strength increases rapidly as the composition approaches stoichiometry. This similarity suggests the possibility of mode softening occurring in Nb_3Sn, also. Using the proximity-gap inversion analysis[32] on earlier Nb_3Sn tunneling data[33] do show evidence[34] for such mode softening. The analysis including such an increase of $\langle\omega\rangle$ with disorder gives a milder decrease of $N^b(0)$ of only 30% and some reduction of $\lambda/N^b(0)$. However, $\langle I^2\rangle$ remains constant. Therefore both methods of analysis lead to the same conclusion of no variation of $\langle I^2\rangle$ with T_c in Nb_3Sn. Lacking quantitative tunneling results on V_3Si, in Fig. 4(c) we show only the results of critical-field data analyzed[21] by assuming a constant $\langle\omega\rangle$. A rather large $\omega_0 \sim 24$ meV is used in this analysis as suggested from the phonon spectrum in the neutron work.[29] The very slight reduction of λ, however, with the very substantial decrease of $N^b(0)$ with disorder leads to an increase of $\lambda/N(0)$ or $\langle I^2\rangle$ to be a factor of 1.8 as large. As mentioned earlier, the reduction of the resolution in determining ω_0 from $2\Delta/k_BT_c$ in the weak-coupled case[25] can lead to subsequent uncertainty in determining λ from this ω_0 and T_c. For example, ω_0's of 18 and 24 meV both give nearly the same $2\Delta/k_BT_c$, yet, if the smaller $\omega_0 \sim 18$ meV is used for the initially ordered V_3Si, a nearly constant value of $\lambda/N^b(0)$ could result and both $\langle\omega^2\rangle$ and $\langle I^2\rangle$ have to increase proportionally as T_c is reduced.

As for the even more metastable $A15$ compounds Nb_3Ge, which is not actually shown in Fig. 4, both the heat-capacity[4] and critical-field[6,9] measurements give a low $N^b(0) \sim 4.0$ states/(eV spin unit cell). The latter experiment of Ghosh and Strongin[9] also showed the evidence of a large decrease of $\lambda/N^b(0)$ by 50% as T_c is reduced from 20.9 to 11.0 K. This observation, coupled with the strong-coupling feature[31,35] of Nb_3Ge found in the tunneling experiments, suggests a similarity between Nb_3Ge and Nb_3Al. Thus the systematic variations in the microscopic parameters of Nb_3Ge are expected to behave very similarly to those of Nb_3Al shown in Fig. 4(a).

Although one may question the decomposition of the electron-phonon coupling parameter λ into factors of $N^b(0)$, $\langle I^2\rangle$, and $\langle\omega^2\rangle$ in such complicated systems as the $A15$ compounds, the systematic trends shown in Fig. 4 are very suggestive. From the stable $A15$ compounds V_3Si and Nb_3Sn to the nearly stable Nb_3Al and on to the metastable Nb_3Ge, $T_c^{\max}$ increases from 17.2 to 23.2 K, but $N^b(0)$ contrarily becomes smaller. Thus the ratio $\lambda/N^b(0) \propto \langle I^2\rangle/\langle\omega^2\rangle$ progressively increases, although for any individual material introducing disorder leads to either an increase or a constant or even a decrease of $\lambda/N^b(0) \propto \langle I^2\rangle/\langle\omega^2\rangle$. These results are in contrast to the correlation $\lambda \propto N^b(0)$ previously obtained[36] among $V_3Si_{1-x}Ga_x$ $(0 \leq x \leq 1)$ by chemical substitutions and do not support the picture where $N^b(0)$ plays the only essential role.

It is believed that the very strong electron-phonon interaction, as characterized by the large $\langle I^2\rangle/\langle\omega^2\rangle$, may lead to the structural instability or phase decomposition,[37] as been seen in the Pb-Bi system.[38] Such a possibility agrees with the systematic trend observed in going from V_3Si to Nb_3Sn to Nb_3Al and Nb_3Ge. In addition a tendency toward structural instability appears to correlate with the increase of the residual resistivity $\rho(T_c)$ with $\langle I^2\rangle/\langle\omega^2\rangle$ that we have observed among these $A15$ materials for a given composition. It might be further argued that the degree of difficulty in preparing homogeneous high-T_c materials should increase due to this tendency toward instability and lead to a limitation on the achievable T_c. The saturation behavior of T_c observed recently in Nb_3Al near the stoichiometric composition[30] could be understood as an expected outcome of such, although alternative explanations have been proposed.[30]

The inevitably large resistivities in all $A15$ Nb_3Al made to date presumably prevent an experimental confirmation of the resistive lifetime broadening model and an unambiguous determination of $N^b(0)$ for well-ordered, stoichiometric Nb_3Al. Lacking data on clean samples [i.e., with $\rho(T_c) < 50\ \mu\Omega$ cm] in both the radiation damaged[9] Nb_3Al and the Nb-Al cases, any extrapolation of $N^b(0)$ vs $\rho(T_c)$ data to $\rho(T_c) \sim 0$ is largely arbitrary. Consequently the possibility of a high $N^b(0)$ in very clean Nb_3Al is not ruled out. It is clear, however, that such a high $N^b(0)$ comparable to Nb_3Sn is not solely responsible for the extrapolated high $T_c > 21$ K, which presumably must also involve the changes in $\langle I^2\rangle/\langle\omega^2\rangle$ discussed above. On the other hand, the presence of such a peak would be consistent with band-structure calculation.[39] The measured low $N^b(0)$'s in all Nb_3Al samples would then result from the smearing of the density of states with the large resistivities, as suggested by the resistive-lifetime broadening model.[6,27,28]

It is important to recognize that the present analyses are carried out within the conventional framework of the strong-coupled electron-phonon theory of superconductivity which neglects many potential real-metal effects. For example, the formal (Eliashberg) theory is derived upon the assumption of the constant density of states near the Fermi level $\sim \omega_D$. Recently, Pickett and Klein[40] have generalized the Eliashberg strong-coupling theory by including the fine structure in the density of states and come to the conclusion that T_c (of Nb_3Sn, for example) is much

less sensitive to the fine variation in $N^b(0)$ due to inherent averaging processes than is widely appreciated. Moreover, the possible effects of spin fluctuations from the electron-electron interaction have been evoked[41] to describe self-consistently the critical fields of V_3Ga. These results also suggest that T_c should be affected, not only in V_3Ga itself but in Nb_3Sn and V_3Si as well. If the suggested spin fluctuations are included in the analysis for the microscopic parameters of these materials, the result is an increase in λ (electron-phonon) and decrease in $N^b(0)$ for high-T_c V_3Si, V_3Ga, and Nb_3Sn. Although these many-body corrections can change the values of the microscopic parameters, our initial estimates indicate that they would not affect the conclusion that $\langle I^2\rangle/\langle\omega^2\rangle$ gets progressively larger as one goes toward the more unstable materials.

The microscopic parameters of the transition-metal compounds have also been discussed from the theoretical point of view. For instance, Hanke *et al.*[42] suggested that the tendency of forming covalent bonding due to the *p-d* hybridization can lead to a resonancelike increase of the nonlocal dielectric function and to a simultaneous increase of $\langle I^2\rangle$ and a decrease of $\langle\omega^2\rangle$. On the other hand, calculations based on a nonorthogonal tight-binding scheme by Weber and Varma[43] showed the phonon anomaly could be equally well caused by the anisotropic electronic band topologies. Not being in a position to judge the relative merits of these theories we simply note that the former model of Hanke *et al.*[42] does provide a simple physical explanation of the systematic trends of $\langle I^2\rangle$ and $\langle\omega^2\rangle$ observed in Nb_3Al and presumably Nb_3Ge. Also it does not require the existence of a very large $N^b(0)$ to give high-T_c superconductivity. Evidence for the tendency of the formation of the covalent bonds in the metastable $A15$'s has been addressed previously by Tsuei *et al.*[7] and Cargill *et al.*[44]

An alternative pairing mechanism based on the exchange of acoustic plasmons has been suggested by Tutto and Ruvalds[45] and Soukoulis and Ruvalds.[45] Although there is yet no direct experimental evidence supporting or disproving this model, our results, in fact, have shown that high T_c and strong coupling in Nb_3Al and Nb_3Ge can be adequately described within the conventional strong-coupling electron-phonon theory.

In conclusion, in this work we have proceeded from a careful and thorough analysis of the data to obtain the microscopic parameters describing the high-T_c superconductivity in Nb_3Al. A further comparative study including V_3Si and Nb_3Sn shows a more intriguing interplay among the parameters $N^b(0)$, $\langle\omega^2\rangle$, and $\langle I^2\rangle$ of these $A15$ compounds than generally assumed, although, in general, they all exhibit a sensitive degradation of T_c with structural disorder. Moreover, although the conventional strong-coupling superconductivity theory that our analysis is based upon may be subject to further refinements or modifications, it is nonetheless evident that the conventional model, in which a peak in the band density of states near the Fermi level plays the essential role in determining the properties of these superconductors, is not sufficient in general. We conclude therefore that a fully adequate explanation of the systematics of the superconducting properties of the high-T_c $A15$ compounds as a class appears to require other essential ingredients discussed in this paper.

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[1]For a more complete review, see Weger and Goldberg, Solid State Phys. 28, 1 (1973).
[2]R. H. Willens, T. H. Geballe, A. C. Gossard, J. P. Maita, A. Menth, G. W. Hull, Jr., and R. R. Soden, Solid State Commun. 7, 837 (1969).
[3]G. R. Stewart, L. R. Newkirk, and F. A. Valencia, Solid State Commun. 26, 417 (1978).
[4]E. Ehrenfreund, A. C. Gossard, and J. H. Wernick, Phys. Rev. B 4, 2906 (1971).
[5]G. S. Brown, L. R. Testardi, J. H. Wernick, A. B. Hallak, and T. H. Geballe, Solid State Commun. 23, 875 (1977).
[6]H. Weismann, M. Gurvitch, A. K. Ghosh, H. Lutz, O. F. Kammerer, and M. Strongin, Phys. Rev. B 17, 122 (1978).
[7]C. C. Tsuei, S. von Molnar, and J. M. Coy, Phys. Rev. Lett. 41, 664 (1978); and C. C. Tsuei, in *Superconductivity in d- and f-Band Metals,* edited by H. Suhl and M. B. Maple (Academic, New York, 1980), p. 233.
[8]R. Viswanathan and R. Caton, Phys. Rev. B 18, 15 (1978); A. Guha, M. P. Sarachik, F. W. Smith, and L. R. Testardi, Phys. Rev. B 18, 9 (1978).
[9]A. K. Ghosh and M. Strongin, in *Superconductivity in d- and f-Band Metals,* edited by H. Suhl and M. B. Maple (Academic, New York, 1980), p. 305.
[10]J. Kwo, R. H. Hammond, and T. H. Geballe, J. Appl. Phys. 51, 1726 (1980).

[11]J. Kwo and T. H. Geballe, Phys. Rev. B 23, 3230 (1981).
[12]B. Cort, G. R. Stewart, A. L. Giorgi, M. P. Maley, C. L. Snead, Jr., A. R. Sweedler, and S. Moehlecke, Bull. Am. Phys. Soc. 26, 479 (1981).
[13]R. H. Hammond, IEEE Trans. Magn. MAG-11, 201 (1975); see also J. Vac. Sci. Technol. 15, 382 (1978).
[14]W. A. Lanford, R. C. Dynes, J. M. Poate, J. M. Rowell, and P. H. Schmidt, Appl. Phys. Lett. 32, 339 (1978).
[15]A. L. Fetter and P. C. Hohenberg, in *Superconductivity,* edited by R. E. Parks (Marcel Dekker, New York, 1969), Vol. II, p. 866.
[16]T. P. Orlando, J. A. Alexander, S. J. Bending, J. Kwo, S. J. Poon, R. H. Hammond, M. R. Beasley, E. J. McNiff, and S. Foner, IEEE Trans. Magn. MAG-17, 368 (1981).
[17]G. W. Webb, Z. Fisk, J. J. Engelhardt, and S. D. Bader, Phys. Rev. B 15, 2624 (1977).
[18]S. Foner, E. J. McNiff, Jr., B. T. Matthias, T. H. Geballe, R. H. Willens, and E. Corenzwit, Phys. Lett. 31A, 349 (1970).
[19]L. R. Testardi, J. M. Poate, and H. J. Levinstein, Phys. Rev. B 15, 2570 (1977).
[20]N. R. Werthamer, in *Superconductivity,* edited by R. E. Parks (Marcel Dekker, New York, 1969), Vol. I, p. 321.
[21]T. P. Orlando, E. J. McNiff, Jr., S. Foner, and M. R. Beasley, Phys. Rev. B 19, 4545 (1979).
[22]D. Rainer and G. Bergmann, J. Low Temp. Phys. 14, 501 (1974).
[23]N. F. Masharov, Sov. Phys. Solid State 16, 1524 (1975) [Fiz. Tverd. Tela 16, 2342 (1974)]; V. Z. Kresin and V. P. Parkhomenko, *ibid.* 16, 2180 (1975) [*ibid.* 16, 3363 (1974)]. The particular results of this paper of importance here are

$$\frac{2\Delta}{k_B T_c} = 3.53\left[1 + 5.3\left(\frac{T_c}{\omega_0}\right)^2 \ln\frac{\omega_0}{T_c}\right]$$

and

$$\eta_{H_{c2}}(T_c) = 1 + \left(\frac{\pi T_c}{\omega_0}\right)^2\left[0.6\ln\frac{\omega_0}{T_c} - 0.26\right].$$

[24]See Ref. 21 for a detailed discussion of the procedures involved.
[25]B. Mitrovic, C. R. Leavens, and J. P. Carbotte, Phys. Rev. B 21, 5048 (1980).
[26]P. B. Allen and R. C. Dynes, Phys. Rev. B 12, 905 (1975).
[27]A. S. Aleksandrov, F. F. Elsein, and M. P. Kazeko, Sov. Phys. Solid State 21, 1181 (1979) [Fiz. Tverd. Tela 21, 2062 (1979)].
[28]L. R. Testardi and L. F. Mattheiss, Phys. Rev. Lett. 41, 1612 (1978).
[29]B. P. Schweiss, B. Renker, E. Schneider, and W. Reichardt, in *Superconductivity in d- and f-Metals,* edited by D. H. Douglas (Plenum, New York, 1978), p. 189.
[30]J. L. Jorda, R. Flukiger, A. Junod, and J. Muller, IEEE Trans. Magn. 17, 557 (1981).
[31]D. F. Moore, R. B. Zubeck, J. M. Rowell, and M. R. Beasley, Phys. Rev. B 20, 272 (1979).
[32]E. L. Wolf, John Zasadzinski, J. W. Osmun, and Gerald B. Arnold, J. Low Temp. Phys. 40, 19 (1980); Gerald B. Arnold, John Zasadzinski, J. W. Osmun, and E. L. Wolf, *ibid.* 40, 225 (1980).
[33]D. F. Moore, M. R. Beasley, and J. M. Rowell, J. Phys. (Paris) 39, C-1390 (1978), Suppl. 8.
[34]J. Kwo, M. R. Beasley, T. H. Geballe, R. H. Hammond, and T. P. Orlando, Bull. Am. Phys. Soc. 26, 211 (1981).
[35]J. M. Rowell and P. H. Schmidt, Appl. Phys. Lett. 29, 622 (1976).
[36]F. Y. Fradin and T. D. Williamson, Phys. Rev. 10, 2803 (1974).
[37]W. L. McMillan, Phys. Rev. 167, 331 (1968).
[38]R. C. Dynes and J. M. Rowell, Phys. Rev. B 11, 1884 (1975).
[39]B. M. Klein, D. A. Papaconstantapoulos, and L. L. Boyer, in *Superconductivity in d- and f-Band Metals,* edited by H. Suhl and M. B. Maple (Academic, New York, 1980), p. 455.
[40]W. Pickett and B. M. Klein (unpublished).
[41]T. P. Orlando and M. R. Beasley, Bull. Am. Phys. Soc. 26, 480 (1981).
[42]W. Hanke, J. Hafner, and H. Bilz, Phys. Rev. Lett. 37, 1560 (1976).
[43]W. Weber, in *Superconductivity in d- and f-Band Metals,* edited by H. Suhl and M. B. Maple (Academic, New York, 1980), p. 131; C. M. Varma and W. Weber, Phys. Rev. Lett. 39, 1094 (1977).
[44]G. S. Gargill, III and C. C. Tsuei, in *Rapidly Quenched Metals III,* edited by B. Canton (The Metal Society, London, 1978), p. 337.
[45]I. Tutto and J. Ruvalds, Phys. Rev. B 19, 5641 (1979); C. M. Soukoulis and J. Ruvalds, J. Low Temp. Phys. 40, 89 (1980).