Comments and Addenda

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Universal defect behavior in Nb-X superconductors: The lattice-parameter correlation

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A correlation between the reduction in T_c and the increase in lattice parameter is shown for a wide variety of chemically diverse, stoichiometric and nonstoichiometric, bulk and film, as-grown, and radiation-damaged niobium-based A-15 superconductors. The correlation is discussed in terms of generalized defects. The T_c of nearly defect-free Nb_3Si is also predicted.

From improved bulk preparation methods, versatile film-deposition techniques, and instructive radiation-damage studies there is now emerging a common characteristic in the family of high-T_c, A-15-structure superconductors. This behavior, which has been previously noted in the literature,[1-3] indicates the existence of a generic structural "fault" which is related to some long-outstanding anomalies of these materials, and the elimination of which is crucial to achieving a high superconducting transition temperature. In this note we point out one of the universal features of this behavior by showing the correlation of the reduction in T_c with the increase in lattice parameter which occurs in a wide variety of Nb-X-A-15 superconductors with different chemical composition and physical properties. The data, which have come from numerous laboratories, has all been published.[1-13] The existence of the correlation is not new; it has been previously observed by some workers in individual compounds. Here we demonstrate its generality, and discuss on this basis the shortcomings of existing ideas, and finally use it to predict the T_c that may be ultimately achieved for Nb_3Si.

In Fig. 1 we have reduced numerous data for the superconducting temperature T_c and the lattice parameter a of a wide variety of A-15 Nb-X samples to a common plot[14] (see Table I for a description and references). For data where a range of T_c and lattice parameters are available we have plotted the reduction in T_c versus the increase in the lattice parameter both relative to the best (highest T_c) sample of that investigation or, in a couple of cases, to the best sample reported elsewhere. The dashed line indicates the average behavior only. The negative curvature for small $\Delta a/a_0$ cannot be justified from the scatter in the plot, but appears to be present in several of the more detailed measurements from which the data has been obtained.

Note that in all cases a reduction in T_c is accompanied by an increase in the A-15 phase lattice parameter. Furthermore, within the admittedly large scatter of the plot,[15] one finds that the correlation is maintained no matter what the source of the data. From Table I it is seen that the correlation includes bulk samples in various forms, CVD, and a wide variety of sputtered films. For Nb-Ge, where large intentional chemical variations were made, the same behavior is obtained either by changes in stoichimetry or at fixed chemical composition. The existence of the correlation for radiation-damaged samples where the composition is held fixed shows that composition is not the intrinsic parameter which determines the reduction in T_c. The different nature of the X atom in the Nb-X compounds also highlights the "universality" of the behavior. We would like to point out that there are exceptions to the correlation, even among binary Nb-X systems (Nb_3Sn, for example); however the large number of diverse cases where the correlation is satisfied leads us to believe that this is more than just an accident.

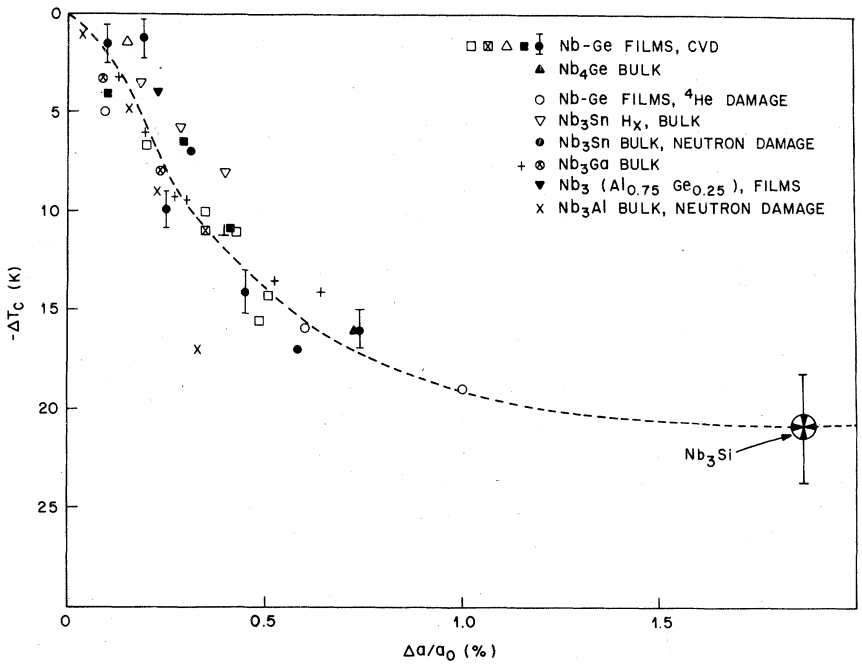

FIG. 1. Reduction in T_c vs lattice-parameter expansion for the A-15 structure Nb-X materials of Table I. See Table I for references to data symbols. Not all data points in the original references have been plotted; however, the points shown are representative of the original results.

Thus, neither stoichiometry (on a macroscopic scale) nor any other identifying characteristic from Table I *per se* is the underlying cause of the correlation. In view of the radiation-damage experiments it appears that the correlation results from a generalized defect which is common to the entire family. This explanation has been previously advanced,[1-3] in a more limited sense, to explain the radiation-damage studies of A-15 superconductors and the resistance ratio versus T_c

TABLE I. Alloys, compositions, and preparation methods for data of Fig. 1.

Symbol	System	Composition	Preparation	References
△	NbGe	~3/1	Sputtered films	Gavaler *et al.* (Ref. 12)
⧳	NbGe	2.6–5.5	Sputtered films	Testardi *et al.* (Ref. 2)
▲	NbGe	~4/1	Bulk	Carpenter and Searcy (Ref. 4), Poate *et al.* (Ref. 3)
○	NbGe	~3/1	Sputtered films plus ^4He damage	Poate *et al.* (Ref. 3)
■	NbGe	2.3–5.7	Sputtered films	Chencinski and Cadieu (Ref. 5)
□	NbGe	~3/1	CVD	Newkirk *et al.* (Ref. 6)
⊠	NbGe	~3/1	Sputtered films	Ghosh *et al.* (Ref. 7)
▽	Nb_3SnH_x	$x \sim 0$–0.6	Hydrogenated bulk	Vieland *et al.* (Ref. 13)
◍	NbSn	~3/1	Bulk tape plus neutron damage	Bett (Ref. 8)
⊗	NbGa	~3/1	Bulk	Johnson and Douglas (Ref. 9)
+	NbGa	~3/1	Quenched and annealed, CVD, arc melted and annealed, I_2 transport	Webb and Englehardt (Ref. 10)
▼	$Nb_3Al_{0.75}Ge_{0.25}$		High-rate sputtered film	Dahlgren (Ref. 11)
×	NbAl	~3/1	Bulk plus neutron damage	Sweedler and Cox (Ref. 1)

correlation of Nb-Ge films.

One type of defect in these materials, which has been studied by a number of workers, is the antistructure defect in which A atoms occupy B sites and vice versa, while retaining the original A_3B structure. Sweedler and Cox[1] have recently used this model to account, with qualitative success, for the lattice-parameter expansion of neutron-irradiated Nb_3Al. Although we believe that such defects will accompany a general type of lattice imperfection, an explanation based on antistructure defects alone is not consistent with all of the existing data.[2,3] In particular T_c does not change with the Nb/Ge ratio in the way expected if the integrity of the Nb atom chains is important for high T_c,[2] and x-ray studies of Nb-Ge films subjected to ^{4}He radiation damage show that the intensity of the high-angle reflection lines become very weak,[3] contrary to the expected result for a perfect lattice with only antistructure defects.

The physical nature of the defect may be, as has been suggested from other evidence,[3,16] the loss of the lattice symmetry as a result of microscopic strains on the scale of the unit cell. The exact microscopic nature of the defect thus may be of secondary importance and perhaps, not even identical for all the samples studied.

The correlation shown in Fig. 1 also shows that the use of the Geller radii to predict the lattice parameters of A-15 compounds entails an assumption about the perfection of the structure which cannot be related to nonstoichiometry alone. For example, "good" (high T_c) Nb_3Sn (bulk) and Nb_3Ge (film) have lattice parameters $a_0 \simeq 5.29$ Å and $\simeq 5.14$ Å, respectively, (see Table I for references). When bulk "Nb_3Ge" was found to have $a_0 \simeq 5.17$ Å the expansion $\Delta a \simeq 0.03$ Å was assumed to be the result of nonstoichiometry. However, Fig. 1 shows that stoichiometric but "bad" (low T_c) Nb_3Sn and Nb_3Ge both have lattice-parameter expansions of about 0.02–0.03 Å. Thus, deviations from the predicted Geller lattice parameters may result as readily from defects, even at fixed chemical composition, as from nonstoichiometry.

One application of Fig. 1 is to predict the T_c of nearly-defect-free Nb_3Si. This can be done by assuming the correlation to hold for this compound, and by noting that the Geller[17] (and Johnson and Douglass[18]) estimates of the lattice parameters (in the low-defect state) are sufficiently accurate for intercomparisons. For Nb_3Si the predicted a_0 is $\simeq 5.08$ Å, while Hammond and Hazra[19] find $a_0 \simeq 5.17$ Å in a sample having $T_c \simeq 9$ K. Thus, with $\Delta a/a_0 \simeq 1.8\%$, Fig. 1 predicts that nearly-defect-free Nb_3Si will have a T_c about 22 K higher than that observed in the data-point sample. This prediction of a $T_c \simeq 30$ K,[20] based on the lattice parameter versus ΔT_c correlation, may be compared with other predictions ($T_c \sim 25$–38 K) obtained by different arguments,[21] and a recent experimental report by Pan *et al.*[22] who obtained $T_c \simeq 19$ K.

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[14] The correlation is mainly derived for binary Nb-X systems, although we have included data points for two ternary compositions. In general, however, addition of a third element can lead to large changes in the lattice parameter due to chemical effects (Vegard's law), which will alter the correlation shown in Fig. 1.
[15] Errors in T_c for this plot are usually dominated by the width of the transition, typically 1–2 K. Errors in $\Delta a/a_0$ have a much greater spread, depending on experiments, and are relatively larger. On the average they are very roughly comparable to the scatter in the plot.
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that $-\Delta T_c$ will be greater than ~16 K, giving $T_c \geq 25$ K.
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