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STABILIZATION OF HIGH- T_c Nb_3Ge *

J.R. Gavalier, M. Ashkin, A.I. Braginski and A.T. Santhanam

Westinghouse Research and Development Center, Pittsburgh, Pennsylvania, 15235, U.S.A.

Résumé.- Les résultats d'analyses physiques nous font croire que la croissance de Nb_3Ge stoechiométrique se fait par homoépitaxie sur une phase A15 de Nb-Ge qui se forme près de l'interface avec le substrat et a une constante de réseau augmentée grâce à la présence des impuretés.

Abstract.- From analytical results we hypothesize that stoichiometric Nb_3Ge is formed through a homo-epitaxial process from an A15 Nb-Ge phase having an impurity expanded lattice formed near the interface.

INTRODUCTION.- High- T_c Nb_3Ge ($T_c \sim 22$ K) can now be formed by low-energy sputtering /1/ chemical vapor deposition (CVD) /2/ and coevaporation /3/. These high- T_c films which crystallize in the A15 structure are close to or have the ideal 3/1 composition, and have a lattice parameter ~ 5.14 Å. Substantial evidence indicates that impurities enter in a beneficial way in forming high- T_c Nb_3Ge /4-6/. However, with no deliberate introduction of impurities, epitaxial growth on Nb_3Ir also produced high- T_c Nb_3Ge films /7/. Thus the role of impurities in forming high- T_c Nb_3Ge remains unresolved.

To try to resolve this question we have been investigating the initially nucleated material in high- T_c sputtered and CVD Nb-Ge films. Based on these results we present a hypothesis which we believe explains the importance of impurities to the formation of the high- T_c Nb_3Ge phase.

EXPERIMENTAL PROCEDURE.- The Nb-Ge films for this study were made by sputtering and CVD according to methods previously described /1,2/. Substrates used were sapphire, Hastelloy B, tantalum and vanadium. In some cases, the tantalum and vanadium substrates were first oxidized by heating in air for several minutes at $\sim 600^\circ\text{C}$. Superconducting T_c 's of the films were determined by a standard four-point resistance technique using germanium thermometry. Composition analyses were done by Auger and Secondary Ion Mass Spectroscopy (SIMS). Structural analyses was by transmission electron microscopy (TEM).

RESULTS.- We have found that in sputtered Nb_3Ge ,

none of the films of 1000 Å or less had the maximum critical temperatures of between 22 and 23 K measured in thicker films. Tarutani and Kudo /8/ have reported similar results for their coevaporated Nb_3Ge films. Chemical analysis of sputtered film has previously shown the presence of oxygen throughout these films with the concentration near the interface being very high (of the order of one oxygen atom per Nb_3Ge unit) and thereafter decreasing with increasing distance from the interface down to a constant level of about 1 at. % in the body of the film. Analyses of CVD films on the other hand showed that there was no oxygen in the body of these films at least down to the 0.1 at. % detection limit of the Auger apparatus used for the analysis. However the analysis did show a sharp oxygen peak at the interface similar to that observed in the sputtered films. To try to determine the significance of this oxygen peak, a series of Nb_3Ge films were sputtered onto alumina, tantalum, and vanadium substrates under similar conditions. Since tantalum and vanadium both have a strong affinity to oxygen, it was reasoned that any oxygen that would be present on these substrates would tend to react with the metal therefore decreasing the possibility of its being incorporated into the initially nucleated films. The results of these experiments showed that whereas the Nb-Ge film deposited onto the alumina substrates had typically high- T_c onsets of ~ 22 K, and widths ΔT_c 's of ~ 1 K, the films on V and Ta had onsets of only about 13 K and ΔT_c 's of 3 to 4 K. The deposition experiments were then repeated using vanadium and tantalum substrates which were first oxidized. With these substrates high- T_c 's could be obtained similar to those on alumina. An oxygen

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profile from SIMS for a high- T_c (~ 21.5 K) film deposited on an oxidized tantalum substrate is similar to all of the other high- T_c films that we have analysed, i.e., the level is highest at the interface and then drops off in the body of the film. This type of profile is not observed in the lower- T_c film deposited on the tantalum substrate; instead the oxygen is low at the interface and increases in the body of the film. These results again point toward the initially nucleated material as being crucial toward achieving high- T_c 's.

To investigate this initially nucleated material transmission electron analyses were made on three very thin (< 1000 Å) sputtered films on sapphire substrates. These films had T_c onsets of 14, 16, and 17 K which were the highest values observed for the particular film thicknesses. The ΔT_c were ~ 2 K. The structure of a representative film with a T_c onset of 14 K and thickness of ~ 700 Å is characterized by extremely fine grains with the grains being clean with no substructure. From the selected area diffraction (SAD) pattern d-spacings were calculated. Most of the observed reflections could be identified as belonging to the A15 Nb_3Ge phase. An unexpectedly large lattice parameter, a_0 , of 5.25 Å was calculated for this phase. Similarly large a_0 's were also found for the other two film studied.

DISCUSSION.— From these observations, we propose the following hypothesis: during the initial ~ 1000 Å of growth a Nb_3Ge phase is formed which is at or close to 3/1 stoichiometry but which has a much larger lattice parameter than is usual for stoichiometric Nb_3Ge (~ 5.25 Å compared to ~ 5.14 Å). We believe that because of this large lattice parameter, this phase having 3/1 stoichiometry is stable and continues to grow without decomposing into a two-phase mixture as would otherwise occur. This concept is consistent with other data on A15 structure superconductors having the general formula Nb_3X (where $X = Sn, Al, Ga, \text{ or } Ge$) wherein the A15 lattice becomes progressively more unstable as the cell size decreases. The boundary between a stable and unstable lattice appears to be when a_0 is ~ 5.17 Å. Continuing with the hypothesis, we believe that as the film growth proceeds by "homoeptitaxy", the A15 lattice gradually becomes smaller until it achieves a cell edge commensurate with its

3/1 composition and with the size of the constituent Nb and Ge atoms, i.e., $a_0 \sim 5.14$ Å. Because of its now small lattice parameter (small Nb-Nb distances) its T_c reaches the very high value of ~ 22 to 23 K. Accepting this picture one can now see that the function of the impurities during the first 1000 Å of growth must be, in some way, to expand the lattice of the stoichiometric Nb_3Ge phase sufficiently to allow it to be stable. One may speculate that this is accomplished by one of the following ways: The presence of a large number of interstitial impurities in the A15 lattice could produce the necessary expansion, or possibly the presence of impurities during nucleation could cause the formation of very fine A15 grains. If these grains were sufficiently small, the lattice could conceivably relax sufficiently to produce the necessary expansion. In either case, as the impurity level decreases, the lattice gradually would be allowed to shrink to its normal dimensions. In the case where one uses an A15 substrate such as Nb_3Ir , the necessity of first forming a stable stoichiometric A15 layer would no longer be required since the epitaxial growth process would commence immediately on the original substrate material. Although at present all of our results appear to support this hypothesis a more complete analysis of a significant number of Nb_3Ge films will be required before one can make a final judgment regarding its validity.

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