Enhancement of the superconducting transition temperatures in ion-implanted aluminium alloys

A.M. Lamoise, J. Chaumont, F. Lalu, Frédéric Meunier, H. Bernas

Classification
Physics Abstracts
7.188 — 8.362

ENHANCEMENT OF THE SUPERCONDUCTING TRANSITION TEMPERATURES IN ION-IMPLANTED ALUMINIUM ALLOYS (*)

A. M. LAMOISE, J. CHAUMONT and F. LALU

Laboratoire René-Bernas du Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse, 91406 Orsay, France

F. MEUNIER

Laboratoire de Physique des Solides (**), Université Paris-Sud, 91405 Orsay, France

and

H. BERNAS

Institut de Physique Nucléaire, Université Paris-Sud, 91406 Orsay, France

(*Reçu le 2 juillet 1976, accepté le 15 septembre 1976*)

Résumé. — La température critique T_c d'alliages Al-C, d'Al-Ge et Al-Si, préparés par implantation ionique à très basse température atteint 4,2 K (C), 7,35 K (Ge) et 8,35 K (Si). Cette dernière valeur de T_c est la plus forte valeur obtenue à partir de l'aluminium. Toutes ces valeurs de T_c correspondent à des concentrations d'éléments covalents de l'ordre de 25 %. Le désordre ne peut, à lui seul, expliquer ces augmentations de températures critiques.

Abstract. — Random alloys of C, Si, and Ge with Al, prepared by ion implantation at liquid helium temperatures, have maximum superconducting transition temperatures T_c of 4.2 K (C), 7.35 K (Ge) and 8.35 K (Si). The latter value is the highest yet obtained for an Al-based alloy. All these values are obtained for covalent element concentrations of $\sim$ 25 %. The effect of lattice disorder is not found sufficient to account for the observed T_c enhancements.

One of the crucial problems in superconductivity relates to the mechanisms by which the superconducting transition temperature (T_c) of a metal may be modified. It is well known that alloying and disordering may produce drastic variations in T_c. The latter effect has been often studied on thin films produced by evaporation techniques [1-3] or by sputtering [4]. In superconductors such as Al, large increases in T_c have been found for films produced by quench-condensation (80 K or 4.2 K) of the metal with non-metals such as O_2, H_2, rare gases, Ge or Si on cold substrates [2, 3, 5]. For pure bulk Al, $T_c = 1.2$ K ; typical values found in this way range from 2.0 to 4.5 K, except for Al-Si and Al-Ge which reach 6.05 and 6.6 K respectively [3] within the framework of Macmillan's theory [6], this effect has been variously ascribed to changes in the phonon spectrum or in the electronic density of states at the Fermi level, or to changes in the amplitude of the electron-phonon interaction. Alternative explanations based on electron-electron interactions via other elementary excitations have also been discussed [7-10]. At present, no overwhelming evidence favours any one of these interpretations.

The microscopic structure and the overall homogeneity of the thin film alloys are basic features in these experiments. The recent use of low temperature implantation techniques is an important development here, since they may produce inherently random phases under well-controlled conditions [11, 12]. We have previously reported T_c increases for Al films implanted with Al, O, He and H [12]. While T_c-enhancements were related to lattice disorder in Al-O and Al-He, the effect of H-implantation was particularly striking because of the high resulting T_c value (6.75 K) and the possibility that an ordered compound had been produced by ion implantation. The resistivity annealing properties were also reported [13]. In the

(*) Work partially supported by DGRST under Contract Nb. 74.7.1083.
(**) Associé au C.N.R.S.

present work, we have studied the critical temperatures of Al based thin film alloys with varying concentrations of the covalent elements C, Si and Ge. We report (i) results on the highest T_c values obtained for these systems; (ii) a study of the correlation between T_c and the annealing temperature for the Al-Si system; and (iii) a study of the influence of lattice disorder on the high T_c samples.

Implantation, resistivity measurements, and film preparation techniques are the same as in our previous work [12]. Thin films (8×0.6 mm^2, thickness 500-1 000 Å) were prepared by evaporation on crystalline quartz substrates from a tungsten crucible in a bell-jar vacuum. During the implantation the target temperature was below 10 K. Implantation energies were ajusted in order to obtain fairly *uniform* samples (e.g., 30 keV and 15 keV Si^+ implantations were performed in a 500 Å thick aluminium film).

Since heavy ions sputter varying amounts of the targets, the film composition after ion implantation had to be determined. The composition of the Al-Ge implanted alloys was estimated from a Rutherford backscattering study of the appropriate sputtering coefficients [14]. For the Si and C implanted samples, the alloy concentration was estimated by assuming that the sputtering ratio of Si on Al was equal to that of Al on Al, and that the sputtering of Al by C was negligible.

Resistivity measurements were performed using a standard four-point probe technique [13]. Implanted samples were repeatedly measured after a succession of Ge, Si and C implantations at varying doses in order to correlate the resistivity just above T_c and the superconducting transition temperature with the concentration of implanted atoms [14].

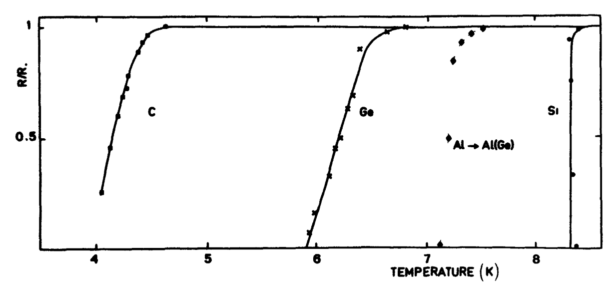

FIG. 1. — Superconducting transitions in Al-C, Al-Si and Al-Ge. The curve indexed Al(Ge) was obtained after Al implantation (dose 7×10^{17} at.cm^{-2}) into a room-temperature co-evaporated $Al_{0.30}Ge_{0.70}$ film. The curve indexed Ge was obtained after Ge implantation (dose 10^{17} at.cm^{-2}) into a pure Al film.

The highest superconducting transition temperature curves obtained for different alloys are displayed in figure 1. They all correspond to estimated average Ge, Si or C concentrations of about 25 %. Note that Al-implanted pure Ge or Al-implanted co-evaporated AlGe films produce higher T_c values than Ge-implanted Al. This could be due to recoil-implanted surface impurities (the forward collision cross-section increases for heavy incoming ions) : the increased concentrations of for example, O, N, or C in the Ge-implanted sample may reduce the maximum T_c value. It is quite possible that the observed value of 7.35 K is still below the maximum obtainable value for Al-Ge.

Three results demonstrate the effect of lattice disorder on the superconducting properties of these alloys : (i) a dose of 10^{16} Si at.cm^{-2} was implanted through the highest-T_c Al-Si sample. The corresponding change in Si concentration was negligible, but the implantation produced ~ 10 displacements per atom in the implanted layer. This had no effect on T_c. (ii) The critical temperature of an Al-Si alloy produced at 6 K was studied after annealing at increasing temperatures (Fig. 2). No drastic variations were found up to about 80 K, but T_c fell to 3.3 K after annealing at room temperature. (iii) The same sample was then Si-implanted at 6 K : T_c was found to increase sharply (Fig. 3) until the average number of displaced atoms in the implanted layer was about one.

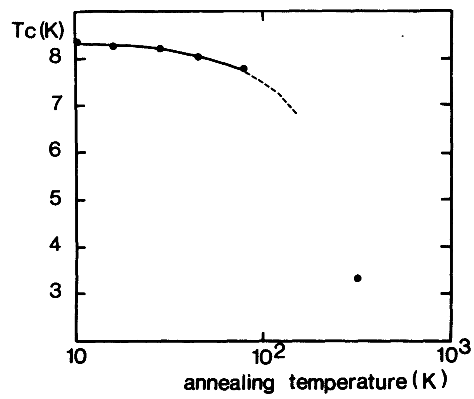

FIG. 2. — Effect of annealing on the superconducting critical temperature T_c of an implanted Al-Si film. All transitions are less than 0.1 K wide.

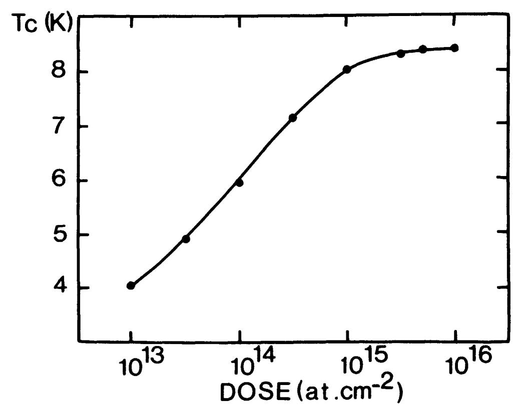

FIG. 3. — Effect of low temperature Si-implantation into Al-Si film. This film was produced by low-temperature implantation, and then annealed at room-temperature. Note negligible composition change : main effect is lattice disordering.

The first result is evidence that the Al-Si alloy produced at 6 K is disordered. Since Al and Si (or Ge) do not form solid solutions, annealing of the implanted samples is expected to result in segregation and crystallite growth : our second result suggests that such a process could take place between 80 K and 300 K [15]. This is corroborated by the third result, since the only possible effect of the low-dose Si implantation is to randomize the atomic distribution in the Al-Si alloy.

The effect of disorder was also witnessed in the Al-Ge system. In several experiments, Al was implanted at 6 K into room-temperature co-evaporated Al-Ge films of varying composition. The implanted Al dose varied from 10^{14} to 10^{18} at.cm^{-2} : in all cases, disordering increased T_c.

Lattice disorder is thus found to favour high T_c-values in Al-Si and Al-Ge. However, a simple interpretation in terms of a softening of the Al phonon spectrum is not satisfying, since the highest T_c-value measured for the equally disordered Al-C, Al-He and Al-O implanted systems [12] is only 4.2 K. From recent experiments on implanted Ge-Cu superconducting alloys [18], it was conjectured that superconductivity is due to a liquid-like metallic phase of the semiconducting component. This suggestion was in line with the fact that superconductivity is obtained on the Ge-rich side of the $Ge_{1-x}Cu_x$ phase diagram (the highest T_c was found around $x \sim 0.3$). In our experiments, however, only Al-rich alloys ($x \gtrsim 0.75$) are superconducting. It is difficult to explain the difference between our results and those of [18] if superconductivity is only due to Ge (or Si). Alternatively, it is interesting to relate the appearance of superconductivity to the metal-insulator transition. This will be discussed for the Al-based alloys in a forthcoming paper [14].

We wish to thank M. Salomé for his help in these experiments. The Rutherford backscattering measurements were carried out at the Groupe de Physique des Solides de l'Ecole Normale Supérieure in collaboration with C. Cohen : it is a pleasure to acknowledge his enthusiastic support and expertise. We are grateful to B. Stritzker and H. Wühl for a preprint of their paper.

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