HAL
open science

Superconducting properties of aluminium thin films after ion implantation at liquid helium temperatures

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

To cite this version:

A.M. Lamoise, J. Chaumont, Frédéric Meunier, H. Bernas. Superconducting properties of aluminium thin films after ion implantation at liquid helium temperatures. Journal de Physique Lettres, 1975, 36 (11), pp.271-273. 10.1051/jphyslet:019750036011027100 . jpa-00231206

HAL Id: jpa-00231206

https://hal.science/jpa-00231206v1

Submitted on 4 Feb 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Classification
Physics Abstracts
8.420

SUPERCONDUCTING PROPERTIES OF ALUMINIUM THIN FILMS AFTER ION IMPLANTATION AT LIQUID HELIUM TEMPERATURES (*)

A. M. LAMOISE and J. CHAUMONT

Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse,
Université Paris-Sud, 91406 Orsay, France

F. MEUNIER

Laboratoire de Physique des Solides, associé au C.N.R.S.,
Université Paris-Sud, 91405 Orsay, France

H. BERNAS

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

(*Reçu le 9 juillet 1975, accepté le 30 juillet 1975*)

Résumé. — La température critique de films minces d'aluminium a été mesurée après implantation d'ions Al, O, He, D et H à des températures inférieures à 6 K, sans réchauffement des échantillons. Les résultats dépendent de la dose et du profil d'implantation, ainsi que des défauts créés. Les limites supérieures des températures critiques obtenues se trouvent entre 2,7 K et 4,1 K après implantation de Al, O et He. Elle atteint 6,75 K pour H, à une concentration moyenne proche de AlH_2.

Abstract. — Using a recent modification of the Orsay ion implantor, Al, O, He, H and D were implanted into Al thin films at temperatures below 6 K. The critical temperature T_c of the implanted films depends on the implanted ion dose and range distribution as well as on radiation damage effects. The higher limit of T_c ranges from 2.7 K to 4.1 K for Al, O and He implantations. It reaches 6.75 K for H, at an average concentration near AlH_2.

It is well-known [1] that the superconducting transition temperature T_c of metals is sensitive to changes in the electron-phonon interaction (hence in the phonon spectrum) as well as in the electronic density of states $N(0)$ at the Fermi surface. In the case of weak-coupling superconductors such as Al, structural changes (disordering) tend to produce fairly large increases in T_c, an effect which is generally ascribed to a *softening* of the phonon spectrum. Specifically, T_c for Al films evaporated on cold ($\sim$ 4.2 K) substrates has been increased from 1.16 K to 3.3 K [2, 3]. Co-evaporation techniques [3, 5] produce films with T_c values up to 6.6 K ; the increase in T_c may not necessarily then be due to disorder alone [5], since such films generally display a granular structure as shown both by X-ray diffraction and very high measured resistivities (ca 10^4 $\mu\Omega$.cm, while the resistivity of cold substrate evaporated Al is typically 10-50 $\mu\Omega$.cm). In this work we have used another method of modifying the phonon spectrum as well as $N(0)$. Ion implantation into Al films at liquid He temperatures produces radiation damage resulting in disorder but solid solutions of pratically any element with the Al host can be produced. In contrast to room temperature experiments [6] the advantage of very low-temperature implantation [7] lies in the fact that neither the implanted impurity nor the related radiation induced defects are mobile. Resistivity annealing experiments can then provide information on migration energies and their relation to T_c [9]. In this work the effect of radiation damage and lattice distortion was studied via Al- and He- ion implantation ; in order to evaluate the possible effect of the recoil-implanted O ions from the surface oxide layer, a study of oxygen implantations was also carried out. A possible influence of optical phonon modes has been underlined by recent experiments on the Pd-H systems [7] : we have performed H- and D-implantations in order to search for similar effects in Al.

In all the cases studied here, ion implantation produced large increases in T_c (in the case of H-

(*) Work partially supported by DGRST Project # 738.

Article published online by EDP Sciences and available at http://dx.doi.org/10.1051/jphyslet:019750036011027100

implantation, we report the highest T_c yet obtained with an Al host). A comparison of the T_c-values, and their relation to the corresponding film residual resistivities leads us to consider : (i) the difference between lattice *distortion* or *amorphization* and lattice disorder due to radiation damage, as well as; (ii) various possible explanations of the rather striking effect of H and D on T_c.

Aluminium films (typically 10 × 0.3 mm^2, thickness 1 300-1 650 Å) were prepared by Al evaporation on crystalline quartz substrates from a tungsten crucible in a standard bell-jar vacuum. Their resistivity ratio ρ (300 K)/ρ (4.2 K) was typically 2.7. Superconducting transition temperatures before implantation were 1.4 K, the transitions being fairly broad (~ 0.7 K) probably due to film edge effects. All ion implantations were carried out with the Orsay ion implantor [8], to which a liquid helium cryostat has been adapted, allowing both low temperature implantation (and implanted ion dose measurements) and resistivity measurements after implantation. With typical ion beam currents of 5-10 μA (1), the estimated target temperature was lower than 10 K; the temperature measured immediately after turning off the beam was $\lesssim$ 6 K on the copper sample holder. Implantation energies were adjusted in order to obtain fairly uniform ion distributions in the host and to avoid proximity effects (the estimated coherence length is ~ 500 Å). Thus for Al-implantation, a single implantation of 50 keV Al^{++} was sufficient; for H-implantation, three implantations were performed at about 10, 5 and 2.5 keV/amu respectively. As discussed in [9], precise values of the implanted sample resistivity were difficult to obtain. However it is certain that the average resistivity of the films did not exceed values between about 10 μΩ.cm and about 50 μΩ.cm for the H-implanted film. The resistivity measurements were performed using a standard four-point probe technique; the temperature was monitored by a calibrated carbon resistor.

For the Al-implanted sample, T_c was measured repeatedly after a succession of 50-keV Al^{++} implantations at varying doses. For the other ions, the same procedure was followed, first at an energy corresponding to *long-range* implantation, then when a maximum T_c was found at an energy corresponding to shorter range implantation (adjusted in terms of the implanted ion profile [10] as well as the coherence length) and so on.

Table I presents our main results on T_c and the superconducting transitions at T_c^{max} are shown in figure 1. The behaviour of the Al-H and Al-D samples is found to be qualitatively different from that of the other implanted alloys. We shall discuss the latter first.

Al-implantation should only produce radiation damage. Using standard collision theory evaluations, we estimate that each atom in the film has been displaced approximately 10 times on the average. This could have been expected to produce *amorphization* of the film and hence increase its resistivity and T_c-value near to that observed for cold-substrate evaporated films (i.e. ~ 3.7 K [3]). Our result does not substantiate this view. Oxygen-implantation was originally undertaken to determine the effect of recoil implantation by Al ions. Considering the results obtained here, the T_c value for Al-implantation could indeed largely correspond to surface oxygen recoil implantation according to a rough estimate based on [11] (it should be noted that due to cryopumping, a number of light atomic species may form a surface layer on our samples and suffer recoil implantation as well). Since chemically inert He produces a very similar value of T_c (albeit at higher doses), it seems unlikely that the increase in T_c has a purely electronic origin. On the other hand, the radiation damage produced by O and — especially — He is far smaller than that due to Al, since light ion slowing-down is mainly due to so-called *electronic* stopping (which

(1) For the high dose H-implantation, ion currents upto 30 μA were used.

TABLE I

Maximum superconducting critical temperatures T_c^{max} measured after ion implantation into Al *thin films. Multiply-charged ion beams or molecular beams were used for pratical reasons. This does not influence the results. For example* 50 keV Al^{++} *ions have the same range and range distribution as* 100 keV Al^+ ; *the same is true, e.g., of* 20 keV H_2^+ *and* 10 keV H^+_{ions}. T_c^{max} *is measured at the middle of the superconducting transition curve.*

Implanted ion	Film thickness (Å)	Implantation energy (keV) (Implanted dose) (at. cm^{-2})			T_c^{max} (K)
—	—	—			—
Al^{++}	1 500		(50, 3×10^{16})		2.6
O^+	1 300	(50, 10^{17})		(25, 5×10^{16})	4.0
He^+	1 300	(16, 10^{17})		(10, 7×10^{17})	3.7
H_2^+	1 650	(20, 10^{18})	(10, 7×10^{17})	(5, 10^{17})	6.75

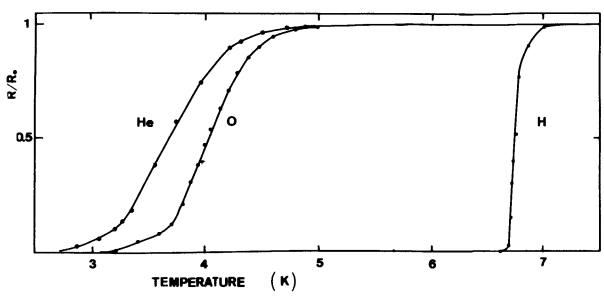

FIG. 1. — Shape of superconducting transition curves for samples producing highest measured transition temperatures.

does not produce atomic displacements). This leads us to suggest that, for O- and He-implanted Al, the increase in T_c is due to the softening of the phonon spectrum via lattice distortion produced by the light ions themselves (as well as by the accompanying *low* damage density) while the highly localized energy deposition of heavier ions (Al) [12] does not necessarily produce the large overall lattice disorder needed to lower the average phonon frequencies. The T_c^{max} value found for O and He could correspond to the *disorder limit* : it would then be far lower than the prediction of [1].

The broad superconducting transitions observed in all these cases can be accounted for on the basis of varying degrees of lattice disorder in the implantation profiles (this is in contrast to the H(D) results discussed below).

Hydrogen and deuterium implantation lead to far higher superconducting transition temperatures and remarkably sharp transitions. This is in contrast to the value (4.1 K) obtained [4] for a co-evaporated, high-resistivity Al-H films. The value of T_c^{max} for H-implanted Al is also higher than that (6.6 K) found in the case of Al-Ge films [5]. It is well known that H(D) is interstitial in metals such as Al, because of its small size and because of charge screening effects (the interstitial nature of H in Al shows up clearly in the resistivity measurements of [9]). This favours lattice distortion, but other effects-related to the electronic density of states or the electron-phonon interaction [1] should also be considered. For example, the electronic density of states could be increased by H(D)-Al bonding (note also possible effects on the Brillouin zone limits) or by the formation of an ordered compound (the average concentration is close to AlH_2). Alternatively, a high concentration of ordered H(D) could lead to an optical phonon branch and thus to an enhanced attraction between electrons due to the electron-optical phonon coupling as was recently found for the Pd-H(D) systems [13].

We cannot now give any conclusion about the isotope-effect : a new experiment is being conducted to study the highest limit of T_c for D-implanted films.

In order to establish the ordered nature of the H(D) distribution in Al, we implanted a fairly low dose (10^{15} at.cm^{-2}) of 50 keV — Al^{++} into the highest — T_c Al-H sample. This dose was presumably just sufficient to displace every atom in the film once on the average. The result of this treatment is shown in figure 2. Clearly, the added disorder *reduces* the superconducting transition temperature down to the limit previously established by O- and He-implantation (and the transition is correspondingly broadened).

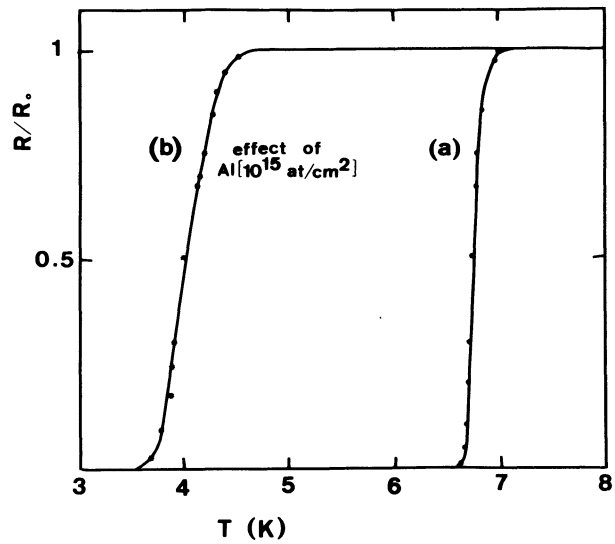

FIG. 2. — Superconducting transition curves for Al-H sample before (*a*) and after (*b*) disordering by a weak Al-ion dose (see text).

If the existence of high T_c values bears more than a chance relation to optical phonon modes, hydrogen-implantation will be an extremely useful tool for systematic investigations of metal-hydrogen alloys.

It is a pleasure to thank Pr. J. P. Burger and J. Friedel and Dr L. Dumoulin for discussions and comments, and F. Lalu, M. Salomé, S. Buhler and the Low-Temperature Group of Institut de Physique Nucléaire, Orsay for their aid in the design, the building and the operation of the cryostat.

References

[1] MC MILLAN, W. L., *Phys. Rev.* **167** (1968) 331.
[2] BUCKEL, W. and HILSCH, R., *Z. Phys.* **138** (1954) 109.
[3] STRONGIN, M., *Physica* **55** (1971) 155.
[4] MINNIGERODE, G. V. and ROTHENBERG, J., *Z. Phys.* **213** (1968) 397 ;
DEUTSCHER, G. and PASTERNAK, A., *Phys. Rev.* **10** (1974) 4042.
[5] FONTAINE, A. and MEUNIER, F., *Phys. Mat. Cond.* **14** (1972) 119.
[6] CROZAT, P., ADDE, R., CHAUMONT, J., BERNAS, H. and ZENATTI, D., *Ion Implantation in Metals*, ed. S. T. Picraux, E. Eer Nisse, and F. Vook (Plenum Press, N. Y.) 1974 ;
MEYER, O., MANN, H. and PHRILINGOS, E., same reference.
[7] STRITZKER, B. and BUCKEL, W., *Z. Phys.* **257** (1972) I ;
STRITZKER, B., *Z. Phys.* **268** (1974) 261.
MACLACHLAN, D. S., MAILFERT, R., BURGER, J. P. and SOUFFACHE, B. (to be published).
[8] CHAUMONT, J., BERNAS, H., SALOMÉ, M., LALU, F., THOMÉ, L., *Vide* **171** (1974) 108.
[9] LAMOISE, A. M., CHAUMONT, J., MEUNIER, F. and BERNAS, H., to be published.
[10] LINDHARD, J., SCHARFF, M. and SCHIOTT, H., *Mat. Fys. Medd. Dan. Vid. Selsk.* **33** (1963) 14 ;
SCHIOTT, H., *Mat. Fys. Medd. Dan. Vid. Selsk.* **35** (1966) 9.
[11] MOLINE, R. A., REUTLINGER, E. and NORTH, J. C., *Atomic Collisions in Solids*, ed. S. Datz, B. Appleton and C. D. Moak (Plenum Press) 1975.
[12] SIGMUND, P., *Appl. Phys. Lett.* **25** (1974) 169.
[13] ROWE, J. M., RUSH, J. J., SMITH, H. G., MOSTOLLER, M. and PLOTOW, H. E., *Phys. Rev. Lett.* **33** (1974) 1297 ;
EICHLER, A., WÜHL, H. and STRITZKER, B., *Solid State Commun.* **17** (1975) 213.