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Stage II recovery in proton-irradiated aluminium studied by positrons

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Abstract. Defect recovery of low-temperature proton-irradiated aluminium has been studied by the positron lifetime technique. The positron lifetime parameters are observed to change significantly for isochronal annealing in stage I (30–45 K), stage II (45–190 K) and stage III (190–300 K). Stage I is the stage where interstitial atoms become mobile leading to annihilation of vacancies and interstitial atoms. In stage III the loss of vacancies at sinks and the growth of vacancy clusters, due to freely migrating vacancies left over from stages I and II, is observed. In stage II mobile interstitials form three-dimensional clusters that again rearrange into interstitial loops. Unexpectedly, we also observe remarkable changes in the positron lifetime parameters in this temperature region, which is explained by positron trapping at interstitial-type defects. The positron lifetime at these interstitial clusters is less than 230 ps, well below the positron lifetime in a monovacancy, but close to lifetimes deduced from recent studies on deformed aluminium. Comparison with diffuse x-ray scattering results show that the positron lifetime technique is sensitive to both the growth and the structural changes of interstitial clusters.

1. Introduction

Post-irradiation annealing of metals irradiated at low temperatures causes distinct annealing stages, which have been most frequently observed by electrical resistivity measurements (Corbett 1966). The two major stages are the free migration of self-interstitials in stage I (typically 30–60 K) and the free migration of monovacancies in stage III (200–500 K). Stage II is the intermediate region between stages I and III and often contains several substages, some of which are caused by impurities.

The knowledge of the behaviour of self-interstitials in the annealing stage II is important for the overall understanding of the defect recovery in metals. The identification of the various small substages has, however, been difficult since there are only few techniques, which are selectively sensitive to interstitial-type defects.

The diffuse scattering of x-rays has presumably given most information on the behaviour of interstitial atoms in metals (Ehrhart 1984). It has been found that interstitials become mobile in stage I and start to form clusters. At an early stage the clusters are three-dimensional (3D) and collapse into two-dimensional (2D) interstitial loops at

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elevated temperatures. The same behaviour was observed also in a theoretical study (Ingle *et al* 1981), where it was found that for cluster sizes below about 10 atoms 3D clusters are energetically more favourable than 2D clusters, while it is *vice versa* for larger clusters.

The positron annihilation techniques are particularly useful to study open-volume defects, because the positrons seek out and become trapped at such defects. The trapping is reflected by a lowered annihilation rate, due to the lower electron density sensed by the positron in a vacancy-type trap compared to the perfect lattice. The positron lifetime in a monovacancy is approximately 50% longer than in perfect bulk. The lifetime increases further by vacancy clustering into three-dimensional agglomerates and reaches 450–500 ps in large voids (Nieminen and Manninen 1979). Volumes smaller than that of the monovacancy can also trap positrons, especially at low temperatures, and the positron lifetime in such defects can be close to that in the bulk (Nieminen 1983, Linderoth and Hidalgo 1987).

Positron techniques have been applied only in few experiments to study defect annealing after particle irradiation below stage I (Vehanen *et al* 1982, Hansen *et al* 1985, Linderoth *et al* 1987, Rajainmäki *et al* 1988a, b). Stage I gives a clear signal in the positron lifetime parameters since a loss of vacancies reduces the positron trapping probability to vacancies. Previous results on aluminium implanted with protons or helium at 20 K (Linderoth *et al* 1987, Rajainmäki *et al* 1988b) motivated us to make a more detailed study without implanted atoms. Mantl and Trifthäuser (1978) studied the Doppler broadening of the positron annihilation radiation in aluminium after electron irradiation at liquid helium temperature. They observed an anomalous increase of the defect specific line shape parameter around 100 K which they suggested could be due to formation of interstitial clusters.

2. Experimental details

The employed aluminium samples were 6N purity single crystals with thicknesses of 350 μm. The proton irradiations were performed at a low-temperature irradiation facility (Hansen *et al* 1985), where it is possible to keep the specimens continuously below 20 K. The sample-sets were irradiated from both sides in order to obtain as homogeneous a defect profile as possible. In the first set (Al-1) the dose of 1.1×10^{15} protons cm^{-2} was given with the mean energy of 5 MeV. The mean energy is the energy of the protons in the middle of the sample–source–sample sandwich where a major part of the positrons annihilate. After a subsequent lifetime measurement the irradiations were continued with the same proton energy until a dose of 4.5×10^{15} protons cm^{-2} was achieved. Similarly in a second sample-set (Al-2) two irradiations were performed, but with doses of 8.8×10^{15} protons cm^{-2} and 2.2×10^{16} protons cm^{-2}, respectively, and with a mean energy of 9.5 MeV. The initial energies of the proton beams for Al-1 and Al-2 were 9 and 12 MeV, respectively. For Al-2 the proton energy was sufficient to let all protons penetrate the whole sample set (Littmark and Ziegler 1977). For sample set Al-1 the protons stopped in the specimens. However, the fraction of positrons annihilating in this hydrogen-containing region is estimated to be below 2%, and has no significant effect on the defect recovery observed by the positron lifetime measurements.

The isochronal annealings were followed by positron lifetime measurements performed at 15 K using a BaF$_2$-based fast coincidence time spectrometer (Rajainmäki 1987) with a time resolution of 210 ps (full width at half maximum). The time resolutions were determined by measurements on defect-free Al, Mo and Ni samples and analysing the spectra with the Resolution program (Kirkegaard *et al* 1981). The source contribution due to the source-foil (170 ps, 13.5%) was found using the empirical estimates by Linderoth *et al* (1984), while the contribution due to annihilation in the source-salt (450 ps, 2.5%) was obtained from the analysis of the measurements on the defect-free samples of aluminium. The spectra from the irradiated samples were then analysed with the Positronfit program (Kirkegaard *et al* 1981) with the source contributions subtracted. Each lifetime spectrum contained 0.7–1 million counts. The spectra were fitted with both one and two lifetime components. For the irradiated specimens two lifetime components were needed to give a satisfactory variance of the fits. With a one-component fit a large variance was obtained. The average lifetime was deduced from:

$$\tau_{av} = I_1\tau_1 + I_2\tau_2. \qquad (1)$$

The annealings were performed with an annealing rate of 25 K h^{-1}. All annealings were carried out in vacuum (10^{-8} Pa).

3. Results and discussion

3.1. Equilibrium measurements

Before the irradiations positron lifetime spectra were measured at various sample temperatures to study the effect of temperature on the positron annihilation parameters. Only a single lifetime component was found for spectra measured at 15 K and 300 K yielding 162 ps and 163 ps, respectively. It corresponds to positron annihilation in defect-free aluminium.

A lifetime spectrum was also measured at 600 K. At this temperature thermal vacancies are present. Two exponentials were needed for the analysis to get a good fit ($\chi^2/\nu < 1.2$). The longer lifetime component, τ_2, was found to be 246 ps with an intensity of 56%. Previously reported values for the positron lifetime in monovacancies in aluminium has ranged from 240 to 253 ps (Fluss *et al* 1978, Schaefer *et al* 1984, Puska and Nieminen 1983, Boronski and Nieminen 1986). The temperature dependence of the positron lifetime parameters in aluminium has been studied extensively by Fluss *et al* (1978) and Schaefer *et al* (1984). From the positron lifetime parameters one can extract the positron trapping rate to the thermally generated vacancies by using the standard trapping model (West 1979). The trapping rate, κ, is proportional to the vacancy concentration

$$\kappa = \mu_t c_t. \qquad (2)$$

c_t is the vacancy concentration and μ_t is the specific trapping rate. Thus by using values for the specific trapping rate from the literature, one can deduce the vacancy concentration. The positron trapping rate to the thermally generated vacancies extracted from our measurements agree well with the results of Fluss *et al* (1978) and Schaefer *et al* (1984).

3.2. As-irradiated

The positron lifetime parameters in aluminium after low-temperature proton-irradiations were found after each irradiation dose and the dose dependence on the

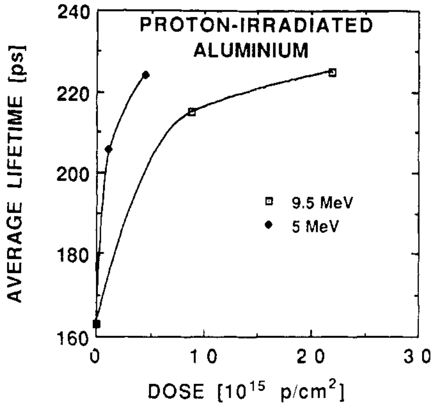

Figure 1. The dose dependence of the average positron lifetime using two different proton energies. The irradiation and measuring temperatures were 20 K and 15 K, respectively. The lines are drawn to guide the eye.

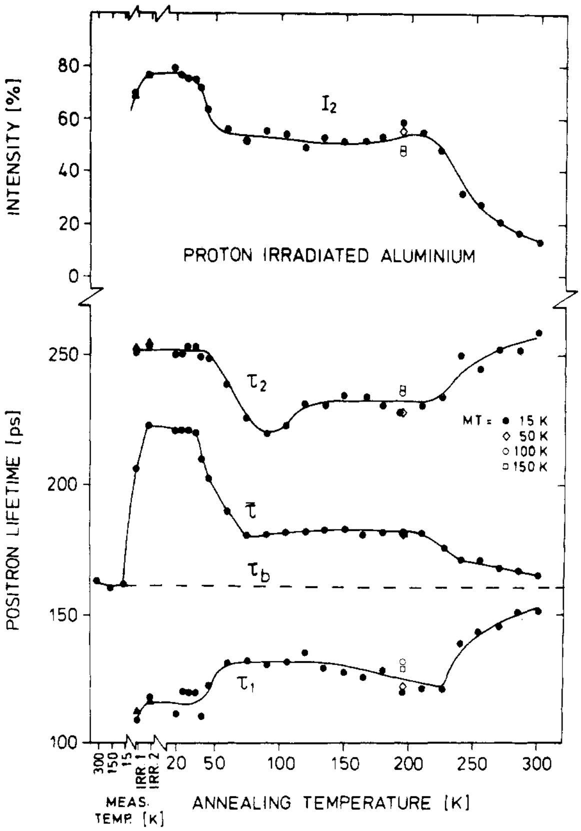

Figure 2. Results of the positron lifetime in parameters 5 MeV proton-irradiated aluminium versus the annealing temperature. The lines are drawn to guide the eye.

average lifetime for both sample sets is shown in figure 1. The average lifetime increased in Al-1 from the bulk value of 162 ps to 206 ps due to the first irradiation and increased further to 224 ps due to the second irradiation. The two irradiations in Al-2 resulted in average positron lifetimes of 215 ps and 225 ps.

Figure 1 shows that damage product rate is higher for 5 MeV protons than for 9.5 MeV protons. This is the result of the energy dependence of the stopping power: the lower the particle energy, the higher the stopping power. Although the stopping power is mainly dominated at this energy range by electronic stopping, also nuclear stopping, the source of the damage, is a decreasing function of particle energy (see e.g. Lehmann 1977). Thus more damage is produced with lower energy particles.

The good time resolution (210 ps, FWHM) and high stability of the spectrometer allowed us to make stable two-exponential fittings. The long lifetime component, τ_2, in Al-1 was 252 ps after both the first and second irradiation, respectively, and in Al-2 the lifetime τ_2 was 254 ps after both irradiations. The positron lifetime in thermally generated monovacancies was found to be 246 ps. The positron lifetime in divacancies is, according to theoretical estimates, some 20 ps longer than in monovacancies (Puska and Nieminen 1983). Thus assuming τ_2 to be an average lifetime for positrons annihilating in mono- and divacancies, it is concluded that 5–10 MeV protons mainly create monovacancies during low-temperature irradiations, but some divacancies are also produced. No significant amounts of larger vacancy clusters are formed, since such clusters would be easily resolvable in the spectrum analysis. With high energy particles one could expect some clusters to be formed because high recoil energies are present. However, we do not observe such clusters which shows that, either only separated vacancies and divacancies are formed in the displacement cascades or that the formed clusters immediately recombine during the collision process.

3.3. *Isochronal annealings*

The defect recovery of aluminium after low-temperature proton-irradiations was studied for both sets of samples and the results are shown in figures 2 and 3.

An annealing stage is seen between 35 K and 50 K for both Al-1 and Al-2. The average positron lifetime decreases from 225 ps to 200 ps and from 224 ps to 200 ps, respectively. The decrease of the average positron lifetime is seen from figures 2 and 3 to be due, solely, to a decrease in I_2, i.e. a decrease of the positron trapping rate to vacancies. This stage is also seen by resistivity and diffuse x-ray measurements on electron irradiated aluminium (Ehrhart and Schilling 1973) and by resistivity measurements on deuterium-irradiated aluminium (Herschbach and Jackson 1967) and it is ascribed to stage I. The decrease of the trapping rate is a result of the recombination of vacancies and migrating interstitials, i.e. the vacancy concentration decreases.

Between 50 K and 120 K one can see two reactions, reflected by first a decrease of the average positron lifetime followed by an increase. This phenomenon cannot be explained by simple reactions between vacancies and interstitials, e.g. where only annihilation of pairs of vacancies and interstitials takes place, since τ_2 first decreases and then increases. Part of the decrease around 50 K in τ_2 can be interpreted as a decrease of the fraction of divacancies. However, the decrease of τ_2 continues below the value of the positron lifetime in a monovacancy (~245 ps), and thus other trapping centres (with a corresponding positron lifetime below 245 ps) must have been formed.

The reactions between 50 K and 120 K must be interpreted as stage II behaviour. After stage I three-dimensional interstitial clusters start to grow (see below) and these clusters are from our measurements found to trap positrons at 15 K. The lifetime associated with such interstitial clusters could be close to the 'bulk' value. It will be included in τ_2 or shared by the τ_1 and the τ_2 components in the two component analysis, resulting in a decrease in τ_2. The distortion of the lattice around the interstitial cluster

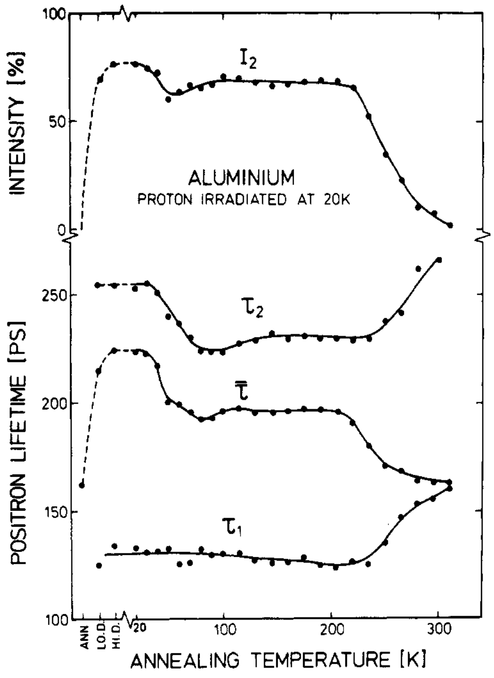

Figure 3. The positron lifetime parameters in 9.5 MeV proton-irradiated aluminium versus the annealing temperature. The lines are drawn to guide the eye.

is presumably small and, hence, the resulting positron binding energy and the deviation from the 'bulk' positron lifetime will also be small. Shallow positron traps with binding energies 0.009 eV and with positron lifetimes as in the bulk have indeed been observed in Ag (Linderoth and Hidalgo 1987). τ_2 in figure 2 reaches a minimum of 220 ps, which shows that the positron lifetime, for positrons trapped at the proposed 3D interstitial clusters, is below 220 ps.

When 3D interstitial clusters contain more than 10 atoms they are found, in theoretical studies, to transform into 2D interstitial loops (Ingle *et al* 1981). This transition occurs around 80 K in electron-irradiated aluminium (Ehrhart 1984). In electron-irradiated aluminium vacancies and interstitials are distributed more homogeneously than in proton-irradiated ones, where separated 'clouds' of vacancies and interstitials are formed. This difference does not seem to affect significantly the stage II behaviour. In the lifetime data the transition in the interstitial cluster geometry is mainly reflected in τ_2, which starts to increase above 80 K. This increase can either be due to 3D clusters containing more traps than loops do or that the positron lifetime in loops is longer than in 3D clusters. τ_2 increases until an annealing temperature of 120 K is reached and then remains constant at about 230 ps until 220 K. These results show that the positron lifetime at the proposed 2D interstitial loops is below 230 ps.

An interstitial loop can be thought of as a chain of 'jogs' (edges in the dislocations), which are known to trap positrons with lifetimes close to the monovacancy lifetime (Smedskjaer 1983, Bentzon *et al* 1985, Hidalgo *et al* 1987). The lifetime, $\tau_2 = 230$ ps, above 120 K suggests that the positron lifetime in a jog is below 230 ps, i.e. shorter than in a vacancy. In fact, in very recent experimental (Hidalgo *et al* 1989) and theoretical (Häkkinen *et al* 1989) studies the positron lifetime at jogs in aluminium has been found to be 215–225 ps.

The temperature dependence of the lifetime spectrum was investigated after the 190 K annealing (see figure 2). Some decrease in I_2 and some increase in τ_2 were observed, but the changes are small because both vacancies and jogs are deep traps for positrons.

In the second sample-set (figure 3) the drop in I_2 around 50 K is followed by a small increase. This implies an increase in the number of trapping centres. This again can be explained by the formation of interstitial loops with larger specific positron trapping rates than the 3D clusters.

At 220 K I_2 starts to decrease rapidly, because vacancies become mobile (stage III) and annihilate at interstitial loops and grain boundaries. The vacancy recombination with interstitial loops also starts to break-up the loops. The increase of τ_2 above 240 K is probably due to formation of small vacancy clusters, since τ_2 exceeds the lifetime in mono- and divacancies. The concentration of these clusters is, however, very small.

4. Summary

Aluminium has been irradiated by 5–10 MeV protons at 15 K. The defect recovery during the isochronal annealing was monitored by the positron lifetime technique. Annealing stages I, II and III were clearly observed. The results strongly indicate that at 15 K positrons are trapped by interstitial-type defects with a lifetime of less than 230 ps; well below that at monovacancies (245 ps).

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References

Bentzon M D, Linderoth S and Petersen K 1985 *Positron Annihilation* ed P C Jain, R M Singru and K P Gopinathan (Singapore: World Scientific) p 485
Boronski E and Nieminen R M 1986 *Phys. Rev.* B **34** 3820
Corbett J W 1966 *Electron Radiation Damage in Semiconductors and Metals* (New York: Academic Press)
Ehrhart P 1984 *Microstructural Characterization of Materials by Non-Microscopic Techniques* ed N Hessel Andersen *et al* (Roskilde: Risø National Laboratory) p 239
Ehrhart P and Schilling W 1973 *Phys. Rev.* B **8** 2604
Fluss M J, Smedskjaer L C, Chason M K, Legnini D G and Siegel R W 1978 *Phys. Rev.* B **17** 3444
Häkkinen H, Mäkinen S and Manninen M 1989 *Europhys. Lett.* **9** 809
Hansen H E, Rajainmäki H, Talja R, Nielsen H K, Nielsen B and Nieminen R M 1985 *Appl. Phys.* A **36** 81
Herschbach K and Jackson J J 1967 *Phys. Rev.* **158** 661
Hidalgo C, Linderoth S and de Diego N 1987 *Phys. Rev.* B **36** 6740
Hidalgo C, Linderoth S, Gonzales G and San Juan J 1989 *Positron Annihilation* ed L Dorikens-Vanpraet, M Dorikens and D Segers (Singapore: World Scientific) p 371
Ingle K W, Perrin R C and Schober H R 1981 *J. Phys. F: Met. Phys.* **11** 1161
Kirkegaard P, Eldrup M, Mogensen O E and Pedersen N J 1981 *Comput. Phys. Commun.* **23** 307
Lehmann Chr 1977 *Interaction of Radiation with Solids and Elementary Defect Production* (Amsterdam: North-Holland)
Linderoth S, Hansen H E, Nielsen B and Petersen K 1984 *Appl. Phys.* A **33** 25

Linderoth S and Hidalgo C 1987 *Phys. Rev.* B **36** 4054

Linderoth S, Rajainmäki H and Nieminen R M 1987 *Phys. Rev.* B **35** 5524

Littmark U and Ziegler J F 1977 *Range Distributions for Energetic Ions in all Elements* (New York: Pergamon Press)

Mantl S and Triftshäuser W 1978 *Phys. Rev.* B **17** 1645

Nieminen R M 1983 *Positron Solid-State Physics* ed W Brandt and A Dupasquier (Amsterdam: North-Holland) p 359

Nieminen R M and Manninen M J 1979 *Positrons in Solids (Topics in Current Physics 12)* ed P Hautojärvi (Heidelberg: Springer) p 145

Puska M J and Nieminen R M 1983 *J. Phys. F: Met. Phys.* **13** 333

Rajainmäki H 1987 *Appl. Phys.* A **42** 205

Rajainmäki H, Linderoth S, Hansen H E and Nieminen R M 1988a *J. Phys. F: Met. Phys.* **18** 1109

Rajainmäki H, Linderoth S, Nieminen R M, Hansen H E and Bentzon M D 1988b *Phys. Rev.* B **38** 1087

Schaefer H E, Gugelmeier R, Schmolz M and Seeger A 1984 *Microstructural Characterisation of Materials by Non-Microscopic Techniques* ed N Hessel Andersen *et al* (Roskilde: Risø National Laboratory) p 489

Smedskjaer L C 1983 *Positron Solid-State Physics* ed W Brandt and A Dupasquier (Amsterdam: North-Holland) p 597

Vehanen A, Hautojärvi P, Johansson J, Yli-Kauppila J and Moser P 1982 *Phys. Rev.* B **25** 762

West R W 1979 *Positrons in Solids (Topics in Current Physics 12)* ed P Hautojärvi (Heidelberg: Springer) p 111