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^{181}Ta TDPAC studies in proton implanted Nb

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Abstract. Time differential perturbed angular correlation (TDPAC) measurement performed on the 482 keV level of ^{181}Ta probe nuclei in *p*-implanted NbHf sample shows that hydrogen decorated vacancy complexes are trapped by 6% of probe nuclei experiencing a Lorentzian distribution of quadrupole frequencies with a width of 32 MHz having a mean value of 545 ± 10 MHz with an asymmetry parameter 0·32. Measurements at RT following isochronal annealing treatment of the sample shows that the onset of detrapping of H–V complexes from probe nuclei occurs at 373 K and gets completed at 573 K. Hydrogen atoms stabilized vacancy clusters are found to be trapped by probe nuclei. The role played by other gaseous impurities in the sample in stabilizing the lattice defects bound to probe nuclei is discussed, to have a complete understanding of the recovery of defects in the sample.

Keywords. Hydrogen-vacancy complex; electric field gradient; defect recovery.

1. Introduction

BCC metals are of technological importance in fusion reactor environments where a detailed knowledge of property changes due to lattice damage is crucial. The presence of impurities like H, O, N and C in these metals influence the defect recovery stages in them. Hydrogen of about 20 appm concentration present in Nb is reported to shift the temperature for migration of monovacancies to 340 K from 240 K which corresponds to that of the metal with negligible hydrogen concentration (Hautojarvi 1987). Lattice defects in a cubic metal result in lowering of local symmetry. The resulting electric field gradient interacts with the nuclear electric quadrupole moment of a suitable probe atom introduced in the sample and causes a time dependent perturbation of angular distribution of γ rays emitted in cascade by probe atom (Pleiter and Hohenemser 1982; Recknagel *et al* 1983). Each defect gives rise to a unique set of well defined modulated frequencies which can be measured with high resolution enabling discrimination between defects occurring simultaneously in the sample (Wrede *et al* 1986). The experiment aims at the following: (i) to study the proton implantation induced defects as trapped by fractions of Hf impurities in Nb, and the evolution of the former bound to hydrogen associated defects, with isochronal annealing treatment of the sample and (ii) to identify other impurities like oxygen, as bound to probe nuclei with or without the association of lattice defects and their evolution with the annealing treatment of the sample.

2. Experimental

Nb Hf sample of 200 μm thickness with 0·6 at% of Hf was neutron irradiated at CIRUS reactor, BARC, to a fluence of 2×10^{18} n/cm^2 to produce probe nuclei in the sample by

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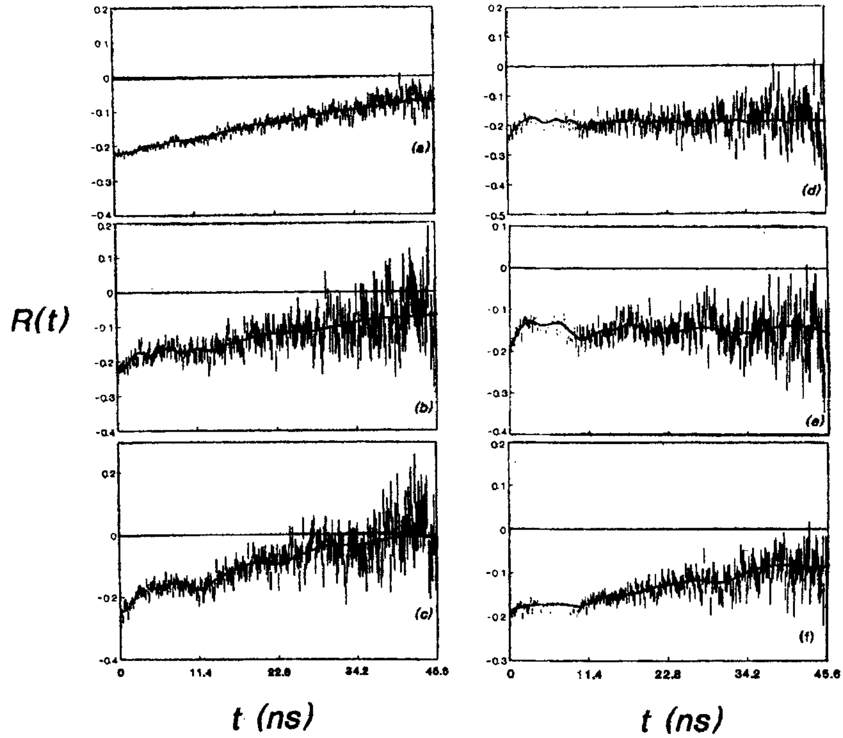

Figure 1. $R(t)$ vs t spectra in NbHf sample subjected to the following treatments: (a) well annealed, (b) proton implanted, (c) $T_a = 423$ K, (d) $T_a = 523$ K, (e) $T_a = 623$ K and (f) $T_a = 723$ K.

the reaction ^{180}Hf (n,γ) ^{181}Hf$\rightarrow\beta^-$ ^{181}Ta. The TDPAC of the 133–482 keV $\gamma-\gamma$ cascade of ^{181}Ta was measured by a three detector twin fast–slow coincidence set up having NaI(Tl) detectors with a prompt time resolution of 1·8 ns FWHM. The sample was subsequently p-implanted to a dose of 2×10^{17} p/cm^2, to a mean depth of 0·6 μm in the sample using a 150 keV linear accelerator of Materials Science Division of our centre. PAC experiments were carried out on the sample at RT and after each isochronal annealing treatment of the sample from 323 K to 823 K in steps of 50 K at a pressure of 10^{-6} torr. The resultant anisotropy spectra for a few annealing temperatures (T_a) are shown in figure 1. The experimentally measured normalized anisotropy spectra $R(t)$ were least squares fitted to the following function (Recknagel *et al* 1983).

$$R(t) = A_2[f_1 \exp[-\delta^1 k_m^1 t] + \sum_{i=2}^{n} G_2^i(t)], \tag{1}$$

$$G_2^i(t) = f_i \sum_{m=0}^{3} a_m^i \exp[-\delta^i k_m^i(\eta^i)\,\omega_Q^i t] \cos[k_m^i(\eta^i)\,\omega_Q^i t], \tag{2}$$

to evaluate the hyperfine interaction parameters corresponding to different fractions (f_i) of probe nuclei such as the quadrupolar frequencies (ω_Q^i), their widths (δ^i) and the asymmetry parameters (η^i). Results for various annealing treatments of the sample are given in table 1.

Table 1. Hyperfine interaction parameters of probe nuclei in proton implanted NbHf.

Frequency (MHz)	Width of frequency (MHz)	Asymmetry parameter	Temp. (K)	Configuration
0	40	0	all	Substitutional Hf
545	32	0·32	300–573	H-V-Hf
167	40	0·12	300–600	H-V_m-Hf
945	80	0·6	above 573	X-V_n-Hf
261	15	0·5	all	Interstitial O–Hf

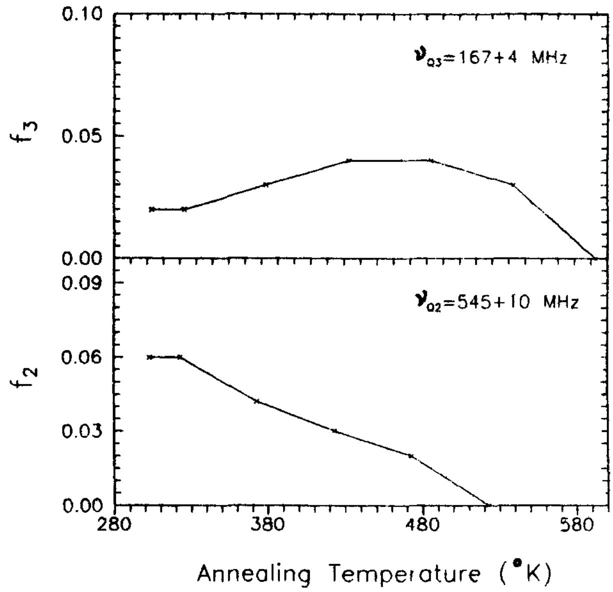

Figure 2. Variation of f_2 and f_3 with isochronal annealing temperature.

3. Results and discussion

Measurement on the reference sample has shown that 0·88 (f_1) fraction of probe nuclei experience a Lorentzian distribution of quadrupole frequencies with a width of 40 MHz having a mean value of 0 MHz. This implies that the above fraction of probe nuclei are substitutional and a large width is due to non-unique en ironment of next nearest neighbour of probe nuclei. The latter could possibly be due to gaseous impurities present in the sample. The remaining fraction of probe nuclei experiences a mean frequency of 261 ± 5 MHz with a Lorentzian width of 15 MHz and $\eta = 0{\cdot}5$. This is due to probe nucleus binding an interstitial oxygen atom, which is in agreement with the results reported earlier (Wrede 1986).

The measurement following p-implantation has shown an appearance of two quadrupole components with parameters viz. $f_2 = 0{\cdot}06$, $\langle \nu_{Q2} \rangle = 545$ MHz, $\eta_2 = 0{\cdot}32$ and $f_3 = 0{\cdot}02$, $\langle \nu_{Q3} \rangle = 167$ MHz, $\eta_3 = 0{\cdot}12$. Variation of fractions of probe nuclei associated with proton implantation induced defects in the sample with annealing treatment is shown in figure 2. The defect configurations are identified to be due to probe nuclei binding $H-V$ complex and $H-V_m$ respectively based on the following: (i) as vacancies are mobile at the temperature of p-implantation, $H-V$ complexes are likely to be trapped by a fraction of probe nuclei, (ii) the above results in the formation of vacancy clusters stabilized by hydrogen atoms that are trapped by probe nuclei, (iii) f_2 starts decreasing beyond an annealing temperature of 350 K which is in agreement with the temperature of dissociation of $H-V$ complex (Thome and Barnas 1978), (iv) the fraction could not either be due to probe nuclei bound to interstitial hydrogen atoms or pure vacancy clusters. If former is the case the value of f_2 will be very large and there may not be any significant variation of it with annealing temperature. Later case is ruled out because of the fact that probe nuclei binding vacancy clusters will experience almost no quadrupole frequency accompanied by an appreciable value of asymmetry parameter. Also we could not find any significant increase in the value of f_1 because of the above and (v) f_3 starts increasing at around 350 K which supports the above interpretation that this fraction of probe nuclei is due to the latter trapping $H-V_m$ complexes.

The defect configuration is completely detrapped from probe nuclei after an annealing treatment at 523 K. About 2% of probe nuclei were found to be associated with a quadrupole interaction frequency of 167 ± 4 MHz with an asymmetry parameter $\eta = 0{\cdot}12$. As this fraction was found to increase to 4%, following annealing of the sample at 373 K, we ascribe this to probe nuclei bound to vacancy clusters decorated by hydrogen atoms. A low value of asymmetry parameter and a large value of relative width of the distribution supports the above interpretation. The formation of this configuration could be due to the dissociation of $H-V$ complex bound to probe nuclei, around this temperature. Annealing of the sample beyond 523 K leads to a decrease in the fraction at the cost of an appearance of a frequency component with $\langle \nu_{Q4} \rangle =$ 945 MHz, $\delta\nu_{Q4} = 80$ MHz and $\eta_4 = 0{\cdot}32$ which is assigned to small vacancy clusters stabilized by gaseous impurities (X) heavier than hydrogen bound to probe nuclei.

4. Conclusions

Hf impurities bind hydrogen decorated vacancy complexes in proton implanted niobium whereas interstitial hydrogen atoms are not bound by the former at room temperature. The measurements following isochronal annealing treatment of the sample show that the onset of detrapping of $H-V$ complex from probe nuclei occurs at 373 K, and gets completed at 523 K. Hydrogen atoms stabilized vacancy clusters are trapped by probe nuclei.

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References

Hautojarvi P 1987 *Mater. Sci. Forum* **15–18** 619
Pleiter F and Hohenemser C 1982 *Phys. Rev.* **B25** 106

Recknagel E, Schatz G and Wichert Th. 1983 in *Hyperfine interactions of radioactive nuclei* (ed.) J Christiansen (Heidelberg: Springer Verlag)
Rinneberg H, Semmler W and Antesberger G 1978 *Phys. Lett.* **A64** 1, 57
Thome L and Bernas H 1978 *Hyp. Int.* **5** 361
Wrede U, Schaefer T and Vianden R 1986 *Z. Phys.* **B64** 461