Chapter 5

HYPERFINE INTERACTIONS STUDIES OF RADIATION INDUCED DEFECTS IN CUBIC Rh METAL.

Contents

5. **Hyperfine interactions studies of Radiation induced defects in cubic Rh metal**

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HYPERFINE INTERACTIONS STUDIES OF RADIATION INDUCED DEFECTS IN CUBIC Rh METAL

5.1 Introduction

Radiation induced defects in metals has been a very important field of study. The induced defects are known to modify the crystalline fields drastically [1]. A cubic metal presents a very conducive environment for the study of defect induced modification in the crystalline fields. The defects-both intrinsic and extrinsic defects create large electric field gradients (EFG) and these EFG can be studied by incorporating radioactive probe nuclei in the vicinity of the defects [2]. Radioactive probe nuclei are incorporated in the matrix by using nuclear reactions and the projectile beam irradiation can cause the radiation damage in the target. Thus the creation of defects and the study of the same is done simultaneously. Hyperfine interactions between the probe-nuclear electro magnetic moments and the defect induced field in the host matrix are very sensitive to the local environment of the nucleus. Through isochronal annealing, the defect dynamics can be studied. Numerous publications report on the annealing behaviour of radiation damage in metals [3,4,5,6,7]. Thus the local probe techniques like Time Differential Perturbed Angular Correlation (TDPAC) and Mossbauer Spectroscopy (MS) can be used with advantage to study such effects.

(*) Dr. K. P. Gopinathan & co workers from IGCAR, Kalpakkam have done very substantial work in this area. It would be good to give at least one reference of theirs.

In this chapter we discuss the effect of radiation damage in cubic (fcc) Rh metal. The nuclear reaction $^{103}Rh(^{12}C, p3n)\ ^{111}Sn\text{----}^{111}In$ with a 69MeV energy , ^{12}C ions from the pelletron of NSC New Delhi, on a cubic Rhodium thick target of 120 μm was used and ^{111}In TDPAC technique was employed to study the local environment around the probe atom.

5.2 Experimental Details

5.2.1 Target Details: The high purity Rhodium (Rh) foil of 1cm x 1cm dimension and thickness of 120 μm was used as the target material. The structure of Rh is close packed FCC and has bond strength of 285 KJ/mole [8].

The sample was mounted on the sample holder of the cryostat. This rectangular shaped cryostat was made of brass which can be filled with Liq. N_2, so that the sample can be cooled to Liq N_2 (77 K) temperature. Whole arrangement was inserted into the vacuum chamber connected to the beam line. The chamber was evacuated upto 10^{-6} torr pressure. This experiment for beam irradiation was done at Material Science Chamber, Nuclear Science Centre, New Delhi. Schematic diagram of beam line setup is shown in Fig. 5.1

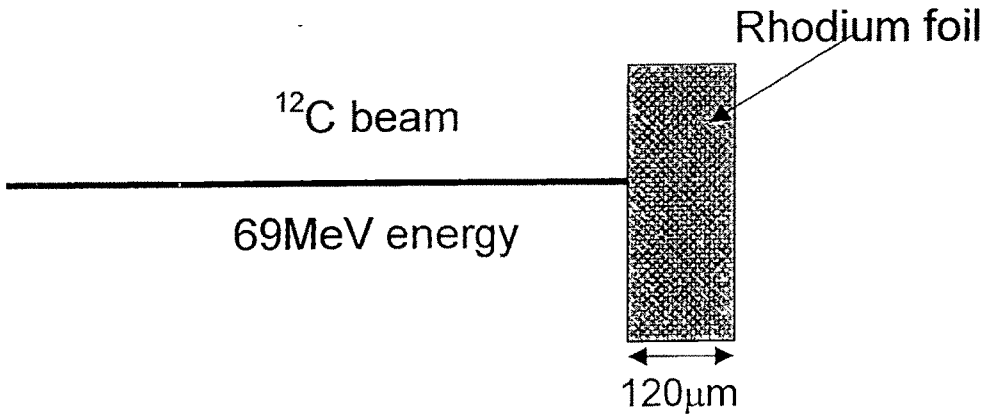

Fig 5.1 Block Diagram of Experimental set up

5.2.2 Irradiation Details:

A beam of ^{12}C with an energy of 69 MeV and charge state +5, was used for nuclear reaction $^{103}Rh(^{12}C, p\ 3n)\ ^{111}Sn$ Maximum cross section of 300 mbarn for the nuclear reaction was calculated from the standard cascade program. The average beam current was 30nA and the size of the focused beam was 0.2cm x 0.3 cm on the target. The target material was irradiated for 8 hours at Liq N_2 temperature. The fluence for 8 hours was calculated as 1 5 x 10^{16} ions/cm^2. The projectile range of Carbon beam is ~ 30μm in Rh, as deduced using TRIM code.

5.2.3 Offline study:

After irradiation, the sample was transferred to another small cryostat containing Liq N_2 which was placed between the detectors used for TDPAC set up. The product nucleus ^{111}Sn so produced during irradiation in the Rh target has the half life ($\tau_{1/2}$) of 35 min, and it decays to ^{111}In via electron capture (EC). This radioactive ^{111}In atom is the TDPAC probe which decays to ^{111}Cd by EC and ^{111}Cd by emitting two successive cascaded gamma rays of 171 and 245 KeV energy. Fig 5.2 shows the decay scheme of the ^{111}Sn produced.

The angular correlation of the 171-245 KeV γ-γ cascade of ^{111}In was measured using a slow - fast coincidence setup comprising of three BaF_2 detectors with twin slow-fast coincidence setup. The time calibration of prompt spectra with ^{22}Na source was calculated as 0.45ns/channel. The plot of calibration is given in Chapter 2. The common detector was gated for the START (171KeV) gamma rays, while other two detectors fixed at 90° and 180° with respect to start detector detected the stop (245KeV) gamma rays. The gating was done through software supported in the Computer Automated Measurement and Control (CAMAC) system used in slow fast coincidence setup. For each position of start detector two time spectra W(90,t) and W(180,t) were recorded simultaneously. From the delayed time resolution spectra W(90,t) & W(180,t), the normalized anisotropy value R(t) was calculated as follows:

$$R(t) = \frac{2[W(180^\circ,t) - W(90^\circ,t)]}{W(180^\circ,t) + 2W(90^\circ,t)},$$

The normalized anisotropy R(t) spectra were least square fitted to a theoretical function of the form

$$R(t) = A_{22}G_{22}(t) = A_{22}\sum_{i=1}^{n} f_i \sum S_{2n} \cos(R\omega(t)) \exp(-R\delta\omega(t)),$$

where $G_{22}(t)$ is the perturbation factor, ω is the precession frequency related to the quadrupole interaction frequency $\nu_Q = (eQV_{ZZ})/h$ and R is a function of $\eta=(V_{xx}-V_{yy})/V_{zz}$. The V_{xx}, etc. are the EFG components in the principal axis system. $\delta\omega=\delta\nu_Q / \nu_Q$ is the distribution in EFG.

To study the radiation induced defects in Rh metal, isochronal annealing of the samples were done at 300K, 1073K and 1473K and the R(t) spectra were recorded respectively at 77 K.

5.3 Results and Discussion

Fig 1 and Fig. 2 shows the spectra of samples at different annealing temperatures. The parameters evaluated are given in Table 1. It can be seen from the table that the as irradiated Rh sample at 77K surprisingly showed a very high quadrupole interaction frequency (ν_{Q1}) of 1150 MHz with a damping of 5% and another

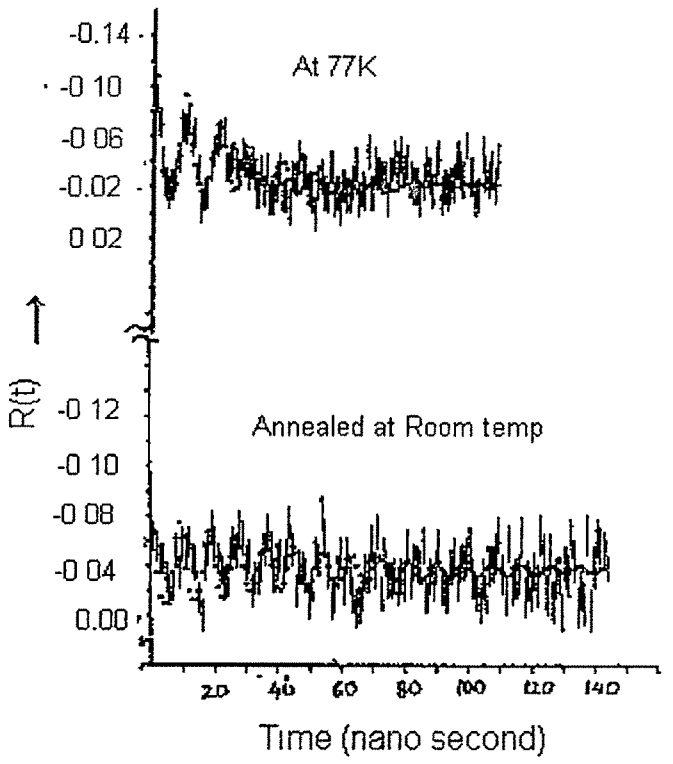

Fig 3 TDPAC Spectra of sample at 77K and 300K

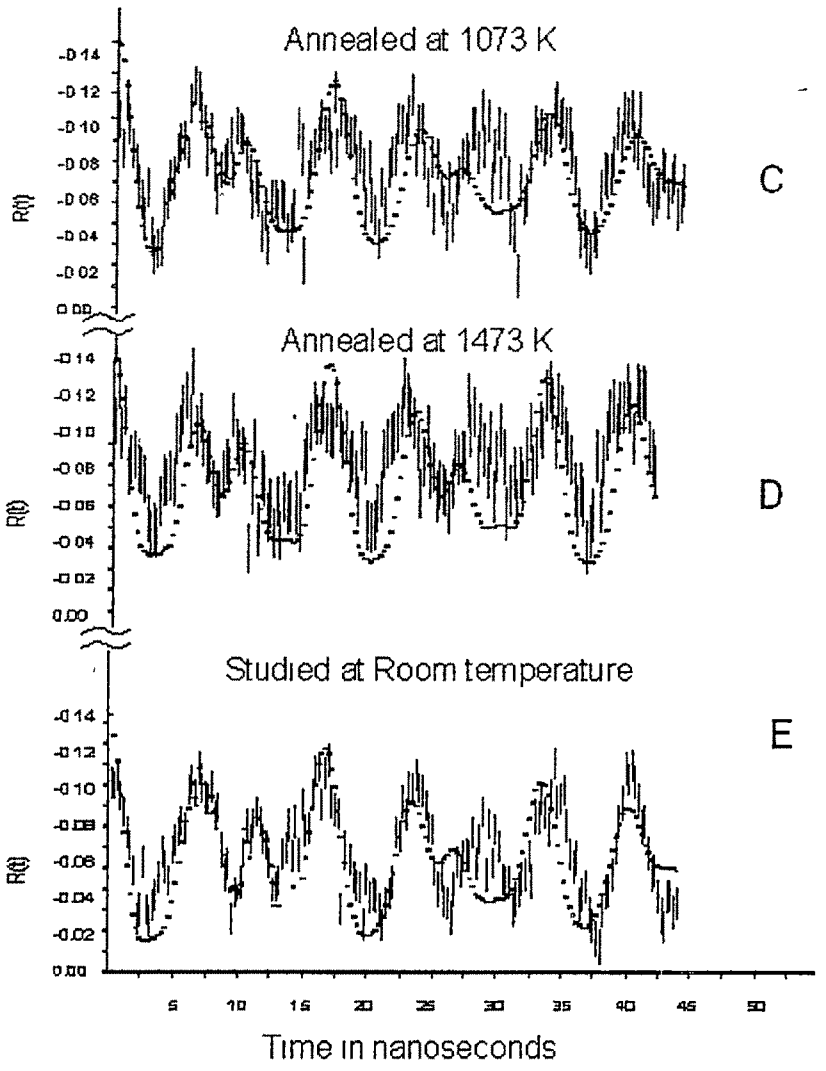

Fig 2 TDPAC spectra of sample at annealing temperatures 1073 K (C), 1473 K (D) Spectra E is studied at Room temperature

Table 1

Annealing Temperatures (K)	ν_{Q1} (MHz)	dω %	η	ν_{Q2} (MHz)	dω %	η
77	1150±10	5	1.00	93±10	10	0
300	1175 ±10	0.9	0.56	--		--
1073	1175±10	0.8	0.56	--		--
1473	1175±10	0.5	0.56	--		--
Sample studied at RT	1175±10	0.4	0.56	--		--

What is the ratio between ν_{Q1} and ν_{Q2}

TDPAC parameters of Rh sample at different isochronal annealing temperatures

After annealing the spectra were taken at R_T. Then what is the meaning of sample studied at R_T ; which is this sample? I must then assume that after annealing, the PAC spectra were taken at 77°k. Is this true?

slow frequency (ν_{Q2}) of 93MHz with a damping of 10%. The asymmetry parameter of ν_{Q1} is found to be 1. Isochronal annealing was done for 10 min at three temperatures 300, 1073 and 1473K. Sample was also studied at Room temperature. The interaction frequency ν_{Q1} at all the temperatures remained same. Also the damping is reduced to negligibly small value (0.4%). The asymmetry parameter (η) remained as 0.56. The second frequency (ν_{Q2}) which disappeared at 300K can be understood as due to ^{111}In trapped at a vacancy formed during irradiation. Since the asymmetry parameter was found to be zero for this frequency (ν_{Q2}), the defect could be a monovacancy or an axial divacancy, as the field gradient is axially symmetric in both these cases. But the higher frequency could be due to In trapped at a complex between Rh and C or Rh and O as observed in other complexes like in oxides. The bond strengths of such possible complexes are known to be very high. But Rh-O system has its decomposition temperature at around 1300K. Since the quadrupole interaction still remains at 1473K, this complex may be ruled out.

However, another possibility could be that of the formation of In-H/Sn-H complexes in the system. In the nuclear reaction ^{103}Rh(^{12}C, p3n) ^{111}Sn indicates that there is a continuous production of hydrogen (Proton) in the matrix along with the product radioactive nucleus ^{111}Sn. But the bond strength of In-H complex is 58 Kcal/mole [7] and hence its dissociation temperature should be low. Thus the possibility of such a complex formation can also be ruled out.

Also at these high fluences (≈ 10^{16} ions/cm^2) during irradiation the amorphisation of the sample may also be considered. But the spectra show well defined frequency without any damping at higher temperatures and so the environment of In seems to be well defined. Hence the probability of amorphisation of the sample seems to be very low. The fact that the anisotropy has increased at higher temperatures indicates more In probes are getting trapped at the defect complexes of Rh-C [8]. This is also understandable in the light of the high diffusion coefficient of In in Rh.

5.4 Conclusions

We have reported here the isochronal annealing behaviour of radiation induced defects in Rh metal. The nuclear reaction $^{103}Rh(^{12}C, p\ 3n)\ ^{111}Sn$-----$^{111}In$ was used for the production of TDPAC probe. From the results, the following conclusions can be drawn.

1. Two frequencies were observed in the as irradiated sample studied at Liq N_2 temperature. The slow frequency of 93 MHz was attributed to In ions trapped at vacancies produced in the radiation damage in the ^{12}C irradiation.
2. The higher frequency remained constant even at the high isochronal annealing temperature(1473k), which could be ascribed to the diffused ^{111}In atoms trapped at Rh-C complexes.

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