RADIATION RESISTANCE OF THERMOSENSITIVE QUARTZ CRYSTAL RESONATORS

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ABSTRACT

Thermosensitive quartz crystal resonators were irradiated by gamma Cobalt60 source and by fast neutrons from Pulsed Neutron Reactor. Resonators are realized on flat parallel quartz plates, yxbl/-10°54'/11°06' crystalline cut operate at 26.5 MHz on 3-td overtone and thickness share C-mode.
It has been measured frequency as function of radiation dose at different temperatures. The offset of temperature frequency characteristic of TSQRs under irradiation of fast neutrons up to 2.10^{14} n/cm^2 and gamma rays up to 5 MRad are investigated.
We can conclude that frequency shift is negligible after irradiation by fast neutrons up to 10^{14}n/cm^2. There is negative frequency shift of about $\pm 8.10^{-6}$ is produced by gamma radiation.

1. INTRODUCTION

Radiation resistance of thermosensitive quartz crystal resonators (TSQRs) is important factor for their application for cryogenic temperature measurement under neutron flounce and gamma environment. To clarify their radiation resistance TSQRs were irradiated by gamma Cobalt60 source and by fast neutrons from Pulsed Neutron Reactor. Neutron radiation was carried out at liquid nitrogen and room temperature and gamma radiation only at room temperature. The frequency of TSQRs is given by:

$$f = \frac{n}{2t}\sqrt{\frac{c'_{66}}{\rho}}$$

[1]

Where **n** is overtone, **t** is thickness of quartz plate, **ρ** is density and $\mathbf{c'_{66}}$ is real elastic module and can be expressed by:

$$C'_{66} = C_{66}\cos^2\theta + C_{44}\sin^2\theta - C_{14}\sin 2\theta$$

Here θ is angle of rotation about X-axis. The elastic modules and the density are the primary "materials" factors that determinate the crystal frequency besides the piezoelectric coupling, resonator contour, electrode and mounting structure.

Cultured Z-growth quartz with up to 30ppm of aluminum and 10ppm of sodium and lithium content has been used. The induced effects of ionizing radiation on quartz crystal resonators can be discussed in terms of a model of one of the primary impurity defects in quartz. This defect is substitutional Al^{3+} defect with an associated interstitial charge compensator, either a H^{+}, Ci^{+}, Na^{+} ion or a hole.
Quartz is grown in an alkali-rich environment and lithium and sodium are trapped interstitially next to the aluminum with the valence electron providing the compensation. The sodium sit off the X-axis in the Z-axis channel; and the resulting Al-Na center couses a strong acoustic loss peak at 53K and a much weaker peak at 135K. Lithium, however, sits on the X-axis and consequently, the Al-Li center shows neither acoustic nor dielectric loss peaks. [Ref.1]
Lopes et al.[Ref.1] discussed the conversion by irradiation of the Al-Li (and Al-Na) centers into a mixture of Al-OH center does not produce an acoustic loss peak. However, acoustic loss peaks at 23K, 100K and 135K are associated with the presence of the Al-hole center.[Ref.1]
Changes in the elastic constants of the crystalline structure, besides causing obvious frequency changes, will also cause changes in the frequency-temperature characteristics of the quartz resonator.

2. EXPERIMENT

The irradiation by gamma rays from Cobalt 60 with intensity of 0,459 kGy/h was carried out at room temperature at INRNE, BAS, Bulgaria. The resonant frequency of TSQRs before and after irradiation at temperature 273K, was measured by step 1MRad. Frequency measurement was made after each dose radiation. Uncertainty of measurement is about $\pm 1.10^{-6}$. The temperature 273K is provided by mixture of ice and water in ratio 2:3 with accuracy of ± 0,01K. Temperature gradient in Dewar is not more than 0,01K/cm.
The irradiation by fast neutrons has been carry out at the pulsed neutron reactor IBR-2, Laboratory of Nuclear Physics, JINR, Dubna, Russia. It produced a full spectrum of fast neutron ranging from 10^{-1}MeV to 20MeV; average energy En≈1MeV. The reactor can deliver the neutron flux up to 10^{12}n/cm^2 over areas up to 20x40cm.
In addition to the neutron, gamma radiation is also produced in the nuclear reaction with maximal dose rates up to 10Gy/s. The average energy is about $E_\gamma \approx 1{,}5$MeV. The flux of the fast neutrons of 10^7-10^8n/cm^2.sec was used for our measurements.
The experimental setup[Ref.2] includes a cryostat for the sensors, nickel foil for both - monitoring of the total reactor power and measuring of the homogeneity of the neutron fluence. The induced activity in this foil was used for monitoring of the total reactor power. To monitor the dose rates, the neutron and gamma dosimeters were mounted around the cryostat. The accuracy to determine the fluence of the fast neutrons and γ-dose was ± 10%. [Ref.2]
The main interest on this setup is its ability to monitor on-line the evolution of the TSQRs by comparing their readout with temperature references that are in principle insensitive to the neutron irradiation.
Setup of measurement of frequency and resistance is shown on Figure 1.

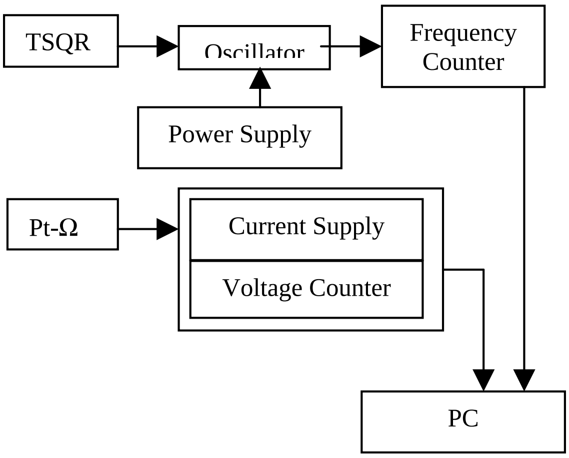

Fig.1. Experimental set-up

A high-frequency matching cable to the measuring oscillator connected the quartz thermosensitive resonator. The frequency was measured by a Ч3-63 frequency counter with uncertainty about of ± $1{,}10^{-9}$. The temperature and frequency data are read every second by the personal computer (PC), where they are displayed as graphics and saved in files.

3. RESULTS AND DISSCUTION

3.1.Gamma radiation

We have measured frequency-temperature curves as functions of radiation dose after each dose gamma rays: 1MRad, 2MRad, 3MRad, 5MRad. Two groups of resonators are irradiated - one with good frequency-temperature characteristic(TFC) and second with bad TFC. Good TFC means that the resonator have smooth TFC without interruption and break of frequency over temperature range from 77K(temperature of liquid nitrogen) to 300K(room temperature). Bad TFC means that the resonator has a lot of interruptions, breaks and its TFC is not smooth.

The frequency depends on gamma dose nonlinearly. There is high frequency shift at low doses up to 2MRad and very small frequency shift at high gamma doses up to 5MRad. There is a saturation of frequency shift after dose 2MRad.

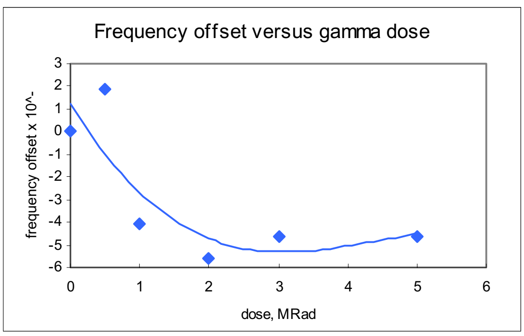

Fig.2. Frequency shift vs gamma dose

3.2. Neutron radiation

We have measured frequency-temperature curves as functions of radiation dose at different temperatures and at different intensity of neutron beam.

Three groups of TSQRs and Platinum resistance sensors were irradiated with the gamma rate 23kRad and neutron flux of 10^{13} - 10^{14} n/cm^2 at room and liquid nitrogen temperatures.

The first group of sensors was putted in liquid nitrogen during irradiation to dose up to $3{,}8.10^{13}$n/cm^2. The second group was mounted on the air close by cryostat where intensity of neutron beam is 7.10^7n/cm^2.sec to dose $5{,}8.10^{13}$n/cm^2. The third group was mounted on the air, near to center of the neutron beam, where its intensity is $1{,}9.10^8$n/cm^2.sec to the dose $1{,}6.10^{14}$n/cm^2.

At the beginning we measured temperature-frequency characteristic of TSQRs and after that the pre- and post- irradiation comparison was made. TSQRs have exhibited the frequency shift Δf/f about $0{,}5.10^{-6}$ during neutron plus gamma radiation doses up to 10^{14}n/cm^2, En>100keV and 2.10^3Gy, Eγ≈1,5MeV at 77,4K and Δf/f about 9.10^{-6} at 300K. These values are within the accuracy of their calibration characteristics and within uncertainty of measurements.

During the test the TSQRs and reference sensors were immersed in the Dewar with liquid nitrogen whose level was kept constant with deviation of ±5mm at the moment of measurement. To avoid the influence of oxygen concentration on saturated pressure of liquid nitrogen, the fresh nitrogen was transferred into the cryostat from the laboratory 1000l storage vessel to minimize contamination. For the presented data the problem with dissolved oxygen were neglected. [Ref.4]

Figure 3 shows frequency shift of TSQR at liquid nitrogen temperature. Stability of the temperature is ±0,01K which corresponding to frequency shift Δf/f about $\pm 10.10^{-6}$, and in our case we obtained Δf/f about $1{,}2.10^{-6}$. This result shows that the frequency shift of TSQR due to irradiation by fast neutrons is in the limits of temperature instability of resonators.

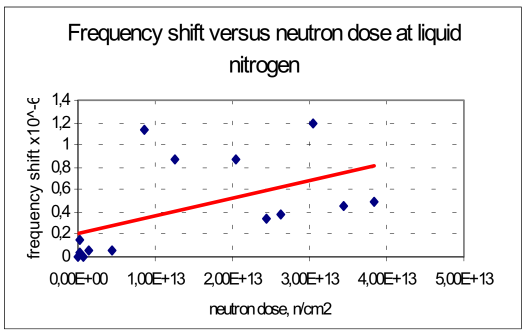

Figure3. Frequency offset versus neutron dose at liquid nitrogen temperature

The Figures 4 and 5 show that frequency shift of TSQRs is the same at different intensity of neutron beam at room temperature.

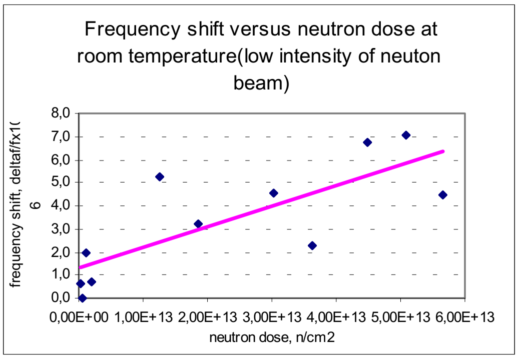

Figure 4. Frequency shifts versus neutron dose of TSQRs on air at middle intensity of neutron beam

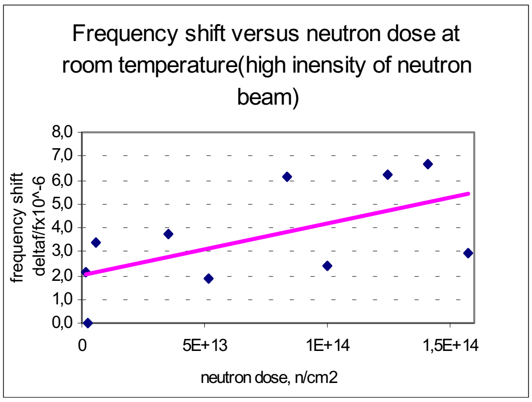

Figure 5. Frequency shifts versus neutron dose of TSQRs on air at high intensity of neutron beam

3.3. Frequency correction procedure

As we want to estimate the irradiation induced frequency offset of our sensors, we must ignore the frequency changes due to the temperature drift. This is done with the following correction procedure:

1) We choose a correction temperature – 77,35K in liquid nitrogen and 290,73K on air.
2) Rejected points – all the points outside the range [77,35 ±0,01]K at liquid nitrogen and [290,73 ±0,2]K on air
3) For each measured point we calculated ΔF=Fmeasured - Fcorrection
4) We obtain: Tcorrect = Tmeasured ± ΔT(dF/dT) where dF/dT is temperature sensitivity of the sensor.

. 5. CONCLUSION

In our case the values of total frequency shift is within accuracy of calibration of TSQRs and within uncertainty of measurement.

We consider that our TSQRs can be used as temperature sensors at cryogenic temperatures under neutron up to the dose 10^{14}n/cm^2 and gamma environment up to 5MRad without frequency shift and offset of temperature-frequency characteristic.

6. REFERENCES

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