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Thermosensitive Quartz Crystal Resonators under Fast Neutrons and Gamma Rays

R. Velcheva[1], **L. Spassov**[1], **Yu. Filippov**[2], **E. Kulagin**[2], **V. Miklayev**[2]

[1]Institute of Solid State Pnysics, Bulgarian Academy of Sciences, 72, Tzarigradsko chaussee Blvd., BG-1784 Sofia, Bulgaria

[2]Joint Institute of Nuclear Research, 141980 Dubna, Moscow region, Russia

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Abstract. Thermosensitive quartz crystal resonators (TSQRs) were irradiated by gamma rays of a Cobalt-60 source and by fast neutrons from a pulsed neutron reactor. Resonators made out of flat parallel quartz plates, $yxbl/10°54'/11°06'$ crystalline cut, operate at 26.5 MHz on 3rd overtone and thickness share C-mode.

The frequency was measured as a radiation dose function for different temperatures. The offset of temperature frequency characteristic of TSQR under fast neutron irradiation of up to 2×10^{14} n/cm^2 and gamma rays up to 5 MRad was investigated. As a result the frequency shift was negligible after irradiation with fast neutrons up to 10^{14} n/cm^2. There was found a negative frequency shift of -8×10^{-6} produced by gamma radiation.

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1 Introduction

Radiation resistance of thermosensitive quartz crystal resonators is an important factor concerning their application in cryogenic temperature measurements under neutron flux and gamma environment. In order to clarify their radiation resistance TSQRs were separately irradiated by both gamma rays from a Cobalt-60 source and fast neutrons from Pulsed Neutron Reactor. Neutron irradiation was carried out at both liquid nitrogen and room temperature, while gamma radiation only at room temperature. The frequency of TSQR is shown as follows:

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

where n denotes the overtone, t is the thickness of quartz plate, ρ is the density and c'_{66} is the real elastic module, which can be expressed by

$$c'_{66} = c_{66}\cos^2\theta + c_{44}\sin^2\theta + c_{14}\sin 2\theta.$$

Here θ is the angle between the Z-axis and the main surface of crystal plate.

Cultured Z-growth quartz with up to 30 ppm of Aluminum and 10 ppm of Sodium and Lithium content was 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 the coupling of a substitutional Al^{3+} defect with an associated interstitial charge compensator, either a H^+, Li^+, Na^+ ion, or a hole.

Quartz is grown in an alkali-rich environment, so Lithium and Sodium are trapped interstitially next to the Aluminum through the valence electron, which provides the compensation. The Sodium sits off the X-axis in the Z-axis channel and the resulting Al–Na center causes a strong acoustic loss peak at 53 K and a much weaker peak at 135 K. Lithium, however, sits on the X-axis, and consequently the Al–Li center shows neither acoustic, nor dielectric loss peaks [l].

Lopes et al. [1] have discussed the irradiation conversion of the Al–Li (and Al–Na) centers into a mixture of Al–OH center, which does not produce an acoustic loss peak. However, acoustic loss peaks at 23 K, 100 K, and 135 K are associated with the presence of an Al–hole center [1].

Changes in the elastic constants of the crystalline structure, besides causing obvious frequency changes, will also cause changes in the frequency-temperature characteristic of the quartz resonator.

2 Experiment

The irradiation with gamma rays from Cobalt-60 with intensity of 0.459 kGy/h was carried out at room temperature at the Institute of Nuclear Research and Nuclear Energy (INRNE), Bulgarian Academy of Sciences. The resonant frequency of TSQR before and after irradiation at temperature 273 K was measured in steps of 1 MRad. Frequency measurement was made after each dose of radiation. The uncertainty of the measurement is about ± 20 mK. The temperature of 273.15 K was provided by a mixture of ice and water in 2:3 ratio with an accuracy of ± 0.01 K. The temperature gradient in the Dewar was less than 0.01 K/cm.

The irradiation with fast neutrons was carried out at the pulsed neutron reactor IBR-2 at the Laboratory of Nuclear Physics, Joint Institute of Nuclear Research (JINR), Dubna, Russia. It produces a full spectrum of fast neutrons ranging from 10^{-1} MeV to 20 MeV with an average energy of $E_n \approx 1$ MeV. The reactor can deliver a neutron flux of up to 10^{12} n/cm^2 over areas of up to 20×40 cm. The experimental setup of fast neutron influences measurements is shown in Figure 1.

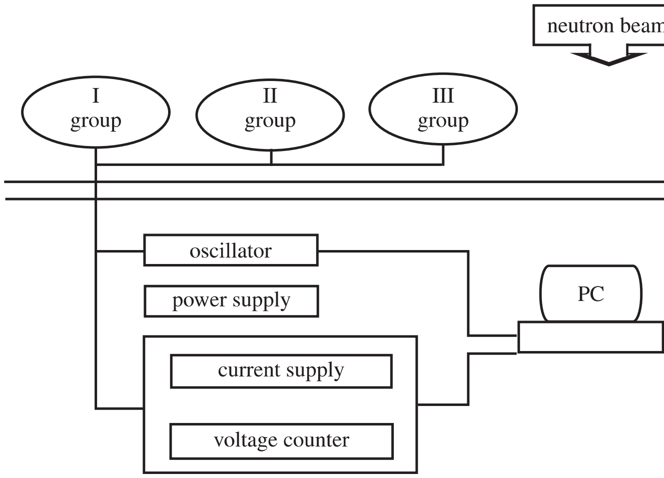

Figure 1. Fast neutron experimental setup.

In addition to the neutrons, gamma radiation was also produced in the nuclear reaction with maximal dose rates of up to 10 Gy/s. The average energy was about $E_\gamma \approx 1.5$ MeV. The fast neutron flux of 10^7–10^8 n/cm^2 sec was used for our measurements.

The experimental setup [2] includes a cryostat for the sensors, nickel foil for both monitoring of the total reactor power and measuring the homogeneity of the neutron flux. The induced activity in this foil was used for monitoring the total reactor power. The neutron and gamma dosimeters were mounted into and around the cryostat to monitor the dose rates. The accuracy of determining the flux of the fast neutrons and γ-dose was $\pm 10\%$ [2].

The main advantage of this setup is its ability to monitor on-line the evolution of the TSQR by comparing its readouts with temperature references, which are in principle insensitive to the neutron irradiation.

The frequency was measured by a CHZ-63 frequency counter with an uncertainty of about $\pm 1 \times 10^{-9}$. The temperature and frequency data were read out every second by a personal computer (PC), where they were displayed in graphical form and saved as files.

3 Results and Discussion

3.1 Gamma radiation

Since the aim of these investigations is the application of TSQRs as temperature sensors working in highly radiated environment, we have investigated the

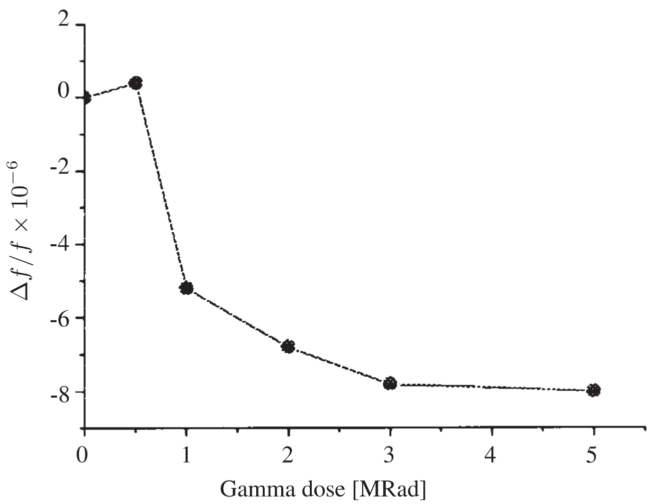

Figure 2. Influence of gamma radiation on the resonant frequency.

influence of radiation on the metrological characteristics of TSQR.

We measured frequency-temperature curves as functions of radiation dose after each dose gamma rays: 1 MRad, 2 MRad, 3 MRad, 5 MRad. TSQRs possess linear frequency-temperature characteristic (TFC) over a temperature range from 77 K (temperature of liquid nitrogen) to 300 K (room temperature) with some nonlinearity from 77 K to 130 K. The investigations show a frequency reduction of -8 $\times$ 10^{-6} after 3 MRad doses gamma radiation. During the consecutive dose increase, the frequency remains constant. The frequency dependence on the gamma dose is nonlinear. There is a high frequency shift at doses below 3 MRad and a very small frequency shift at high gamma doses of up to 5 MRad. There is a saturation of the frequency shift beyond doses of 3 MRad. The positive frequency shift is well visible up to 0.5 MRad. It was possibly caused by relaxation process, surface strain, dissociation, or relaxation effect in electrodes and crystal holders.

3.2 Neutron radiation

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

The TSQRs and reference sensors were immersed during the test in a Dewar with liquid nitrogen, which level was kept constant with deviations of ± 5 mm at the moment of measurement (Figure 1). To avoid any oxygen concentration influences on the saturated pressure of the liquid nitrogen, fresh nitrogen was transferred into the cryostat from the laboratory's 1000 l storage vessel to min-

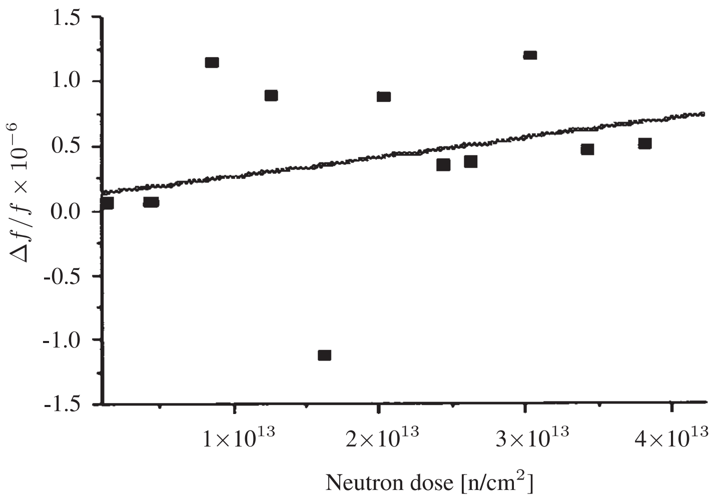

Figure 3. Influence of neutron dose on the resonant frequency at liquid nitrogen temperature.

imize contamination. In the present data, the dissolved oxygen problems were neglected [4].

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

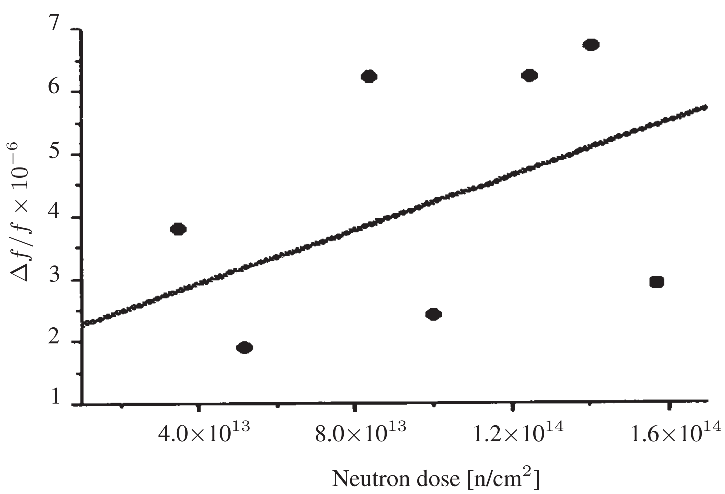

Figure 4. Influence of neutron dose on the resonant frequency at room temperature.

The first group of sensors was placed in liquid nitrogen. It was irradiated up until a 3.8×10^{13} n/cm^2 dose was achieved. The second group was assembled near the cryostat, where the intensity of neutron beam was of 7×10^7 n/cm^{2}sec and dose of 5.8×10^{13} n/cm^2 was achieved. The third group was mounted near the center of the neutron beam, where the intensity was od 1.9×10^8 n/cm^{2}sec and a dose of 1.6×10^{14} n/cm^2 was reached.

In the beginning we measured the temperature-frequency characteristic of TSQR, and then a comparison between pre- and post-irradiation data was made. TSQR showed a frequency shift $\Delta f/f$ of about 1.2×10^{-6} as a result of neutron and gamma radiation doses of up to 10^{14} n/cm^2, $E_n > 100$ keV and 2×10^3 Gy, $E_\gamma \approx l.5$ MeV at 77.4 K and $\Delta f/f$ of about 5×10^{-6} at 300 K. These values are within the limits of the uncertainty measurements of TSQR's calibration characteristics.

3.3 Frequency correction procedure

Since our goal is to estimate the irradiation induced frequency offset of the sensors, we must ignore the frequency changes due to the temperature drift. This was achieved using the following correction procedure:

1) We used adjusted temperatures: 77.35 K in liquid nitrogen, and 290.73 K on air.
2) All points outside the range $[77.35 \pm 0.01]$ K in liquid nitrogen and $[290.73 \pm 0.2]$ K in air were rejected.
3) For each measured point, we calculated $\Delta F = F_{\text{measured}} - F_{\text{correction}}$
4) We obtained $T_{\text{correct}} = T_{\text{measured}} \pm \Delta T(dF/dT)$ where dF/dT was the temperature sensitivity of the sensor.

Figure 3 shows the frequency shift of TSQR at liquid nitrogen temperature. The stability of the temperature is $\pm 0,01$ K, which corresponds to frequency shifts $\Delta f/f$ of about $\pm 10 \times 10^{-6}$. In our case, we obtained $\Delta f/f$ of 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 the temperature instability of resonators.

4 Conclusion

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

We conclude that our TSQRs can be used as temperature sensors at cryogenic temperatures under neutron dose up to 10^{14} n/cm^2 and gamma environment as high as 5 Mrad. There is no frequency shift due to neutron irradiation. There is however an offset of the temperature-frequency characteristic of minus 8×10^{-6} due to gamma radiation.

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