Simple energy level model for frequency degradation of USO under radiation

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***Abstract*— Space Radiations are responsible of transient and permanent shifts of the output frequency signal of any quartz crystal oscillator. In this paper, the degradation of quartz crystals under radiation stresses is shown to be related to competition between trapping and recombination of radiation-induced carriers in quartz lattice. The presented model exhibits a good agreement with experimental data.**

I. INTRODUCTION

Quartz crystal resonators are widely used in satellite communication, guidance systems, positioning applications, temperature and pressure sensors, accelerometers and gyrometers, time-base and ultra stable reference for space applications... In the last named application, the ultra stable oscillators (USO) are usually submitted to ionizing radiations. For example, the trajectory of Jason 1 satellite used for the measurement of sea-floor topography crosses the proton belt in the south Atlantic anomaly (S.A.A.) and more or less large frequency variations are observed [1].

Indeed radiations are responsible to transient and permanent shifts of the output frequency signal of any quartz crystal oscillator. So, an important R&T study managed by the CNES, French space agency, has been started in 2004. Its aim is double:

- to improve the stability of USO under spatial low earth orbit radiations,
- to understand the radiation-matter interaction in quartz crystal material.

Previous studies have shown that radiation sensitivity of quartz oscillator can be predominantly attributed to quartz crystal resonator [2].

In order to explain USO frequency variation under radiation, a simple energy-level model has been considered, with one electron trap and one recombination center. The equations governing the movement of charge carriers created by ionizing radiation are numerically solved. Calculations are made for various parameters to compare our model to experimental data.

Figure 1 shows an example of USO frequency variations for a satellite pass over the S.A.A.

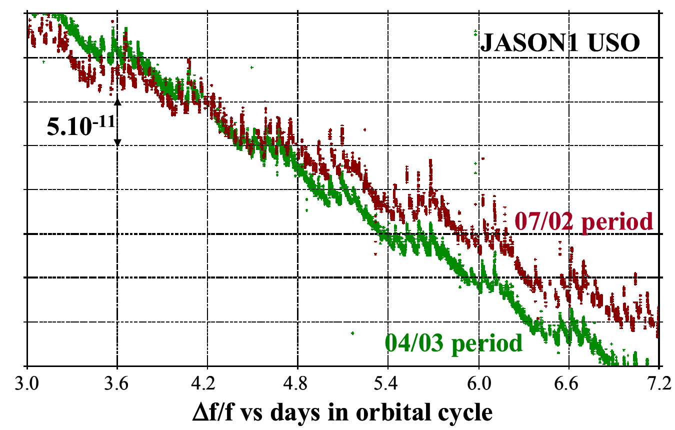

Figure 1. Frequency variations versus time observed on JASON I USO.

II. RADIATION EFFECTS IN QUARTZ

All crystalline quartz contains ubiquitous impurities randomly incorporated during the growth. The dominant impurity in natural and in hydrothermally grown synthetic quartz crystal is aluminium as Al^{3+} cation substituting to Si^{4+}, due to its nearly similar atomic size. An aluminium ion requires additional positive charge compensator, such as Li^{+}, Na^{+}, K^{+} or a proton associated with a non bonding orbital of an adjacent oxygen ion, to compensate the charge of replaced silicon and maintain charge neutrality. The impurity and its compensators give rise to aluminium-hydroxide, aluminium-alkali and aluminium-hole (hole trapped at an adjacent oxygen site) defect centers. The aluminium comes from quartz nutrient (generally natural), while the charge compensator comes either nutrient or mineralizer.

In addition to the Al-related centers, there are other types of point defects also present in quartz: oxygen vacancies, and numerous OH^- molecules formed by protons trapped on oxygen atoms near unidentified point defects in the lattice.

Quartz used in space applications comes from swept material, i.e. alkalii are replaced by hole or proton depending on sweeping environment (air or vacuum). Sweeping quartz in air dissociates Al-alkali and forms Al-OH^-. Sweeping quartz in vacuum produces Al-hole, in addition to the Al-OH^-.

Irradiation produces a large number of uncorrelated electrons and holes pairs which move through the lattice during their short lifetime until they **recombine** or are **trapped**. Protons from OH^- related growth-defects are also released at any temperature, move to aluminium-hole center, replace the holes and rather than remain in ionic interstitial prefer bond to an adjacent oxygen which is associated to aluminium site. Thus, the main effect produced by irradiation of swept quartz at room temperature or above on Al-center is the production a mixture of Al-hole – i.e. trapping of a hole – and Al-OH. Other point defects can trap electron, hole or act as recombination center.

Irradiation of quartz with ionizing radiation leads to an accumulation of trapped charge carriers in defect with energy level within the material's gap.

III. The Model

In quartz there is a wide variety of traps and/or recombination centers for both electrons and holes that could be considered in an energy level scheme. We started with the simplest case: one trap and one recombination center i.e hole trap level where recombination is allowed.

The diagram given in Fig. 2 shows the quantitative energy-level model proposed to study carrier behaviour during and after irradiation. This type of model has been proposed previously to explain dose rate effects in thermoluminescence and optically simulated luminescence experiments [3, 4].

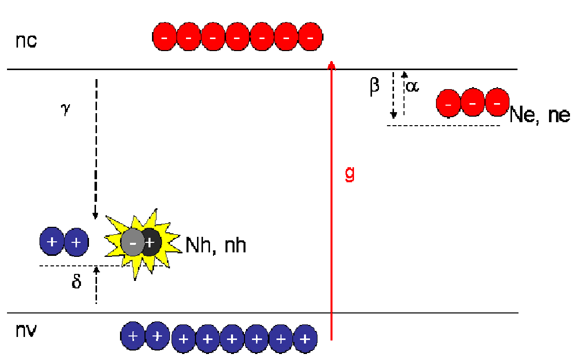

Figure 2. Energy-level diagram used for one eletrcon trap and one recombination center. The arrows shows the transitions allowed. The solid arrow indicates electron-hole pairs production during excitation.

The four simultaneous differential equations governing the trap filling in quartz in the case of one recombination center and one electron trap are:

$$dn_c/dt=g-\beta n_c.(N_e-n_e)+\alpha.n_e-\gamma.n_c.n_h. \quad (1)$$

$$dn_e/dt=\beta.nc(N_e-n_e)-\alpha.n_e. \quad (2)$$

$$dn_h/dt=\delta.n_v.(N_h-n_h)-\gamma n_c.n_h \quad (3)$$

$$dn_v/dt= g-\delta.n_v.(N_h-n_h) \quad (4)$$

The instantaneous concentration of the electrons in the conduction band is noted by $\mathbf{n_c}$ (cm^{-3}), and this of holes in valence bands by $\mathbf{n_v}$ (cm^{-3}). $\mathbf{N_h}$ (cm^{-3}) is the concentration of hole centers with instantaneous occupancy of $\mathbf{n_h}$ (cm^{-3}), $\mathbf{N_e}$ (cm^{-3}) is the concentration of the electron active trapping state with instantaneous occupancy of $\mathbf{n_e}$ (cm^{-3}).

$\boldsymbol{\delta}$ ($cm^{-3}.s^{-1}$) is the trapping probability coefficient for free holes from valence band in center.

$\boldsymbol{\gamma}$ ($cm^{-3}.s^{-1}$) is the recombination probability coefficient for free electrons from conduction band with holes in center, and $\boldsymbol{\beta}$ ($cm^{-3}.s^{-1}$) is the trapping probability coefficient of free electrons from the conduction band into the trapping state.

E (ev) is the activation energy of electron trap.

$\boldsymbol{\alpha}$ (s^{-1}) is the activation factor.

In this system the variables are n_c, n_v, n_h, n_e, the constant are Nv, Ne, and the probability coefficients.

Additional parameter is **g** ($cm^{-3}.s^{-1}$) is the electron-hole pairs rate production which is set to zero after the time of the chosen irradiation.

We assume that band-to-band or trap-to-trap recombination is neglected. We also assume that recombination of holes from valence band and trapped electron is slight.

IV. Numerical Results

Sets of trapping parameters have been chosen and the differential equations were numerically solved during and after irradiation by Runge-Kutta fourth order method using ode45 solver in MatLab to study trap filling. The equations (1)-(4) were first solved for a chosen dose rate and an irradiation time. In order to simulate an orbital cycle, the resolution of the same set of equations is continued for a period of relaxation time with g set to zero.

A. *Center and trap concentration*

High Resolution Glow Discharge Mass Spectrometry (GDMS) is a technique for the analysis of all trace and ultra-trace elemental constituents of inorganic materials. Samples are analyzed in solid form and so do not require the laborious and complicated dissolution methods inherent with such techniques as Inductively Coupled Plasma - Mass Spectrometry. The principle of this technique involves the atomization of a solid sample by sputtering in low-pressure DC plasma. The sputtered atoms are then ionized in the plasma and extracted into the mass analyzer for separation and detection. The Table I presents chemical analyses of three quartz (N= natural, Q1, Q2 synthetic).

TABLE I. GDMS BY SHIVA TECH (PPMA).

	N	Q1	Q2
Al	8.14	1.25	3.85
Fe	<0.18	<0.18	<0.18
Ca	1.15	3.3	8
Na	0.18	0.25	0.86
Li	2.68	0.34	1.74
K	0.27	<0.03	0.72
Ge	<0.14	<0.14	<0.14

One ppma in quartz is about 7.53×10^{16} atoms per centimeter cube. Thus we have chosen electron trap and recombination centers equal to a few 10^{16} per centimeter cube.

B. Comparison between experiment and simulation

We have chosen set of parameters to describe two examples of experimental behaviour of USO under discontinuous γ irradiations. The parameters used for the first simulation correspond in quartz to normal level impurities but with more electron traps than recombination centers. The parameters used for the second simulation are those of a quasi-pure quartz but with more recombination centers than electron traps.

Figures 3 and 5 show experimental data of frequency susceptibility of two USO during discontinuous cycle of irradiation (gamma rays at 1 rad per hour and 4 rad per hour), **Figures** 4 and 6 present the corresponding simulation with our model.

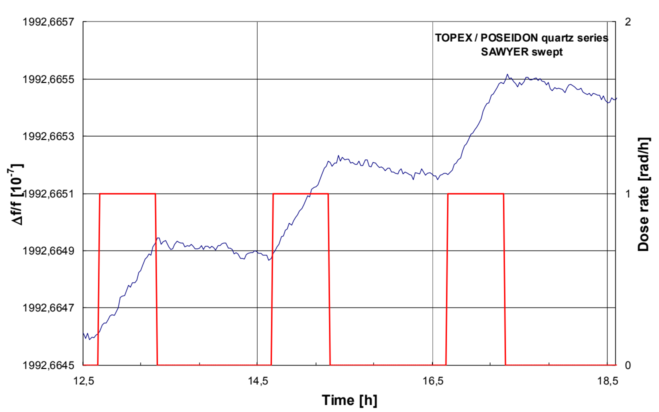

Figure 3. Frequency variations versus time observed during discontinuous γ irradiations (1 rad/hour) (Topex /Poseidon).

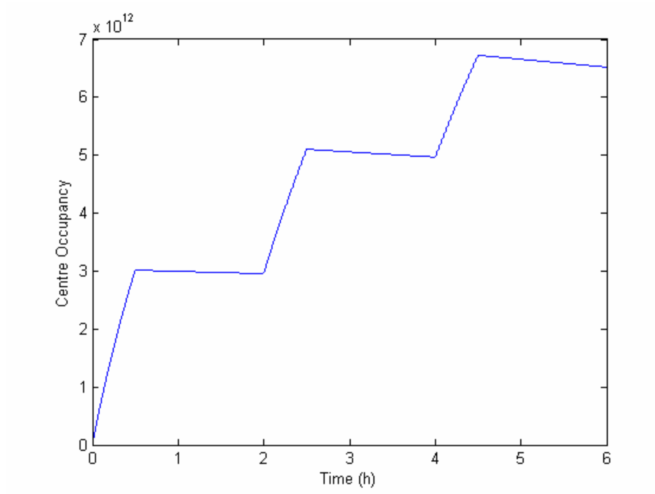

Figure 4. The chosen parameters used in this simulation are the following: Dose rate: 1 rad/h; $N_e=5\times10^{16}$ cm^{-3}; $N_h= 10^{16}$ cm^{-3};$\alpha=10^{-5}$ s^{-1}; $\beta=10^{-17}$ cm^{-3}.s^{-1}; $\gamma=10^{-13}$ cm^{-3}.s^{-1};$\delta=5\times10^{-15}$ cm^{-3}.s^{-1}.

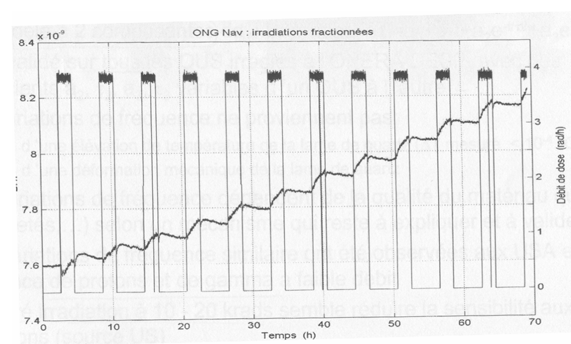

Figure 5. Frequency variations versus time observed during discontinuous γ irradiations (4 rad/hour), (ONG Nav).

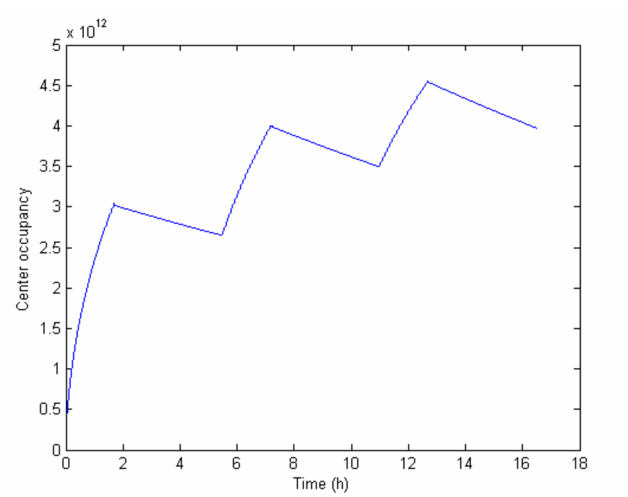

Figure 6. The chosen parameters are used in this simulation are the following: dose rate: 4 rad/h; $N_e=10^{15}$ cm^{-3}; $N_h= 3\times10^{15}$ cm^{-3};$\alpha= 10^{-5}$s^{-1}; $\beta=10^{-17}$ cm^{-3}.s^{-1}; $\gamma= 10^{-13}$ cm^{-3}.s^{-1}; $\delta= 6\times10^{-15}$cm^{-3}.s^{-1}.

V. CONCLUSION

In this work, we have presented a model which can describe the radiation induced frequency shifts for USO submitted to discontinuous irradiations.

Simulations and experiments present the same evolution for various chosen set of parameters. So, with this simple model we show that the behavior of traps filling is the same as those of the USO frequency shift under radiation environment.

Indeed, we know that the level of impurities in quartz is varying from one supplier to another and the typical values are about a few ppma.

ACKNOWLEDGMENT

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