Time Decay of Stable Absorption of Gamma Irradiated Lithium Niobate Crystal Doped by Cuprum Ions

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Abstract. *The present work is devoted to investigation of stability of stable color centers that are induced by gamma radiation in Cu-doped LiNbO3 single crystals*

Key words: lithium niobate, gamma irrdiation, decay of additional absorption.

I. Introduction

Lithium niobate $LiNbO_3$ (LNO) is a ferroelectric crystal having important applications in laser, electrooptic, acoustooptic and optical storage devices [1,2]. The photo-refractive effect, which is used for storage of volume phase holograms, is due to the presence of transition metal impurities (for example Cu) and intrinsic lattice defects [2,3], which are sources and traps of electrons in lithium niobate crystals. That is why study of recharging processes of defects and dopants under external influences (for example ionizing irradiation) is of interest.

The transient and stable color centers in pure and doped LNO crystals have been investigated in several works, for example in [4]. The stable color centers (SCC) are centers which have lifetime higher than 1s. Unfortunately, the lifetime of color centers were studied in literature only for transient color centers, for example in [5]. In this work the lifetime of stable additional absorption induced by gamma irradiation in Cu doped of LNO crystal will be determined from data obtained during 10 years.

II. Experiment

The LNO:Cu (0.03 mol%) crystals were grown from congruent melt in Institute of Electronic Materials Technology (ITME) by the Czochralski technique. Samples for the SCC investigations were made in the form of plane-parallel polished plates of 1 mm thickness. The samples were irradiated with gamma quanta (1.25 MeV) from 60Co with absorbed doses $7*10^5$ Gy. Optical absorption spectra were recorded with SPECORD-M40 and UNICAM 340 spectrophotometers. The value ΔK of additional absorption induced by external influence was determined as

$$\Delta K = \frac{1}{d} ln \frac{T_1}{T_2}$$

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where d is the sample thickness, T_1 and T_2 are the sample transmission coefficients before and after irradiation, respectively.

The additional absorption (AA) was measured three times: immediately after irradiation (04.06.2002), two years after irradiation (03.05.2004) and ten years after irradiation (12.03.2012)

III. Results and Discussions

The "as-grown" absorption spectrum of LNO:Cu crystals is shown in Fig. 1. In LNO:Cu crystals the wide and intensive band centered at 25 000 cm^{-1} is present as well as much less intensive band near 10 000 cm^{-1} is observed. This bands are associated with the intervalent transition $Cu^+ \rightarrow Nb^{5+}$ and $^2E \rightarrow {}^2T_2$ transition in Cu^{2+} ions [6,7], consequently.

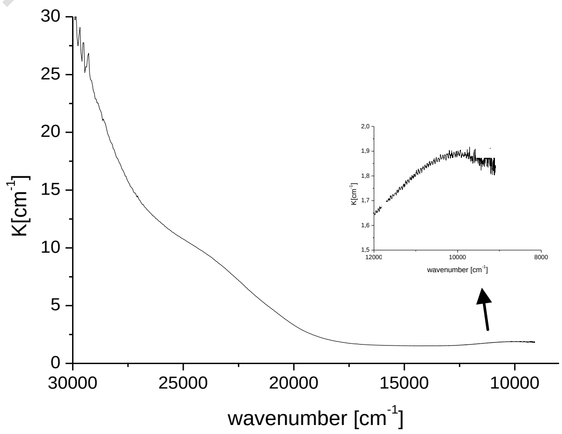


Fig. 1. Absorption spectrum of LNO:Cu(0,03%)

The AA spectrum of LNO:Cu crystal after gamma irradiation represents an intensive wide band with maximum near 25 000 cm^{-1} . Besides, some weak clearing near 10000 cm^{-1} is observed. It was early shown, that such character of AA spectrum indicates that after the annealing of LNO:Cu crystals an increasing of the absorption band caused by Cu^+ ions (at 25 000 cm^{-1}) and decreasing of absorption of Cu^{2+} ions (absorbing at 10 000 cm^{-1}) take place [4]. According literature the contribution in AA with maximum near 25000 cm^{-1} can be due to O^- polarons [8], F^+ [9] or F [10] centers.

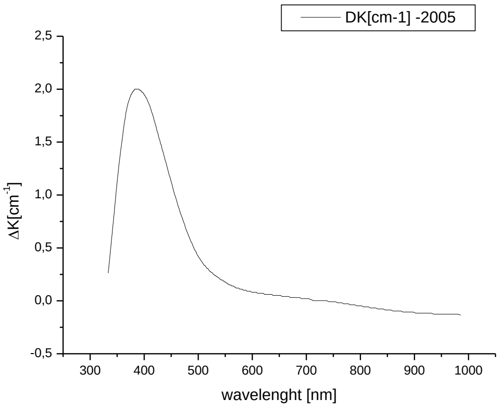
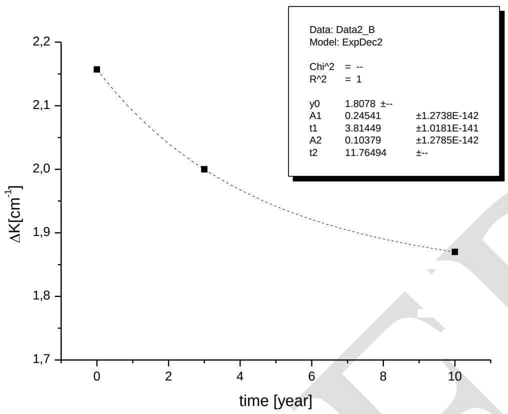


Fig. 2. AA of LNO:Cu measured in 2002 year

Decay kinetic for the AA were measured at 25000 cm^{-1} bands is shown in Fig. 3.



Analysis of this decay kinetic shows that approximation with sum of two exponents is satisfactory. Thus, centers of two types with substantially different lifetimes contribute to the both absorption bands and can be described as:

$$\Delta K(t)_1 = \Delta K_{0,1} + \Delta K_{1,1}\exp\left(\frac{t}{\tau_{1,1}}\right) + \Delta K_{2,1}\exp\left(\frac{t}{\tau_{2,1}}\right) \quad (1)$$

where $\Delta K(t)_1$ is the AA value at time t, $\Delta K_{0,1}$ – AA at the time $t >> \tau_{1,1}$, $\Delta K_{1,1}$ and $\Delta K_{2,1}$ – maximal AA value for the first and second types centers at the beginning of measuring, $\tau_{1,1}$, $\tau_{2,1}$ – lifetimes of the first end second types centers respectively. Numerical values of the equation parameters are listed in Table 1.

To clarify the nature of centers responsible for absorption at 25 000 cm^{-1}, decay kinetic of clearing near 10000 cm^{-1} have been monitored.

Analysis of this decay kinetic shows that approximation with one exponent is satisfactory:

$$\Delta K(t)_2 = \Delta K_{0,2} + \Delta K_{1,2}\exp\left(\frac{t}{\tau_{1,2}}\right) \quad (2)$$

Numerical values of the equation parameters are listed in Table 1.

Table 1. Decay parameters for AA of LNO:Cu crystal

Parameters	Value
$\Delta K_{0,1}$	1,808 $см^{-1}$
$\Delta K_{1,1}$	0,245 $см^{-1}$
$\Delta K_{2,1}$	0,104 $см^{-1}$
$\tau_{1,1}$	3,81 year
$\tau_{2,1}$	11,76 year
$\Delta K_{0,2}$	0,02 $см^{-1}$
$\Delta K_{1,2}$	0,108 $см^{-1}$
$\tau_{1,2}$	9,38 year

It was clear that $\tau_{2,1}$ is approximately equal to $\tau_{1,2}$. According [5], the removing of clearing near 10 000 cm^{-1} can be due to growth of concentration of Cu^{2+} according reaction $Cu^{+} \rightarrow Cu^{2+} + e^{-}$. In this way, the $\tau_{2,1}$ is connected with Cu^{+} ions.

It is interesting that the $\Delta K_{1,1}$ value is about 2.4 times smaller than $\Delta K_{2,1}$. This means that a significant contribution to the absorption with maximum 25000 cm^{-1} may be centers associated with genetic defects

IV. Conclusions

The stable additional absorption decay in LNO:Cu crystal is mainly due to release electron by Cu^{+} ion. The long half-life time of AA is about 12 years.

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