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Dark and Photo-Conductivity Measurement Techniques for Dielectric Materials, Application to $LiNbO_3$

[1,2]B. Chikh-Bled, [1]M. Aillerie and [2]B. Benyoucef
[1]MOPS Laboratory, Paul Verlaine University of Metz and Supelec,
2 rue Edouard Belin, 57070 Metz, France
[2]URMER, University Abou Bakr Belkaid, B.P. 119, 13000 Tlemcen, Algeria

Abstract: Dark conductivity and photo-conductivity are the two mains charge transport phenomena involved in photo-refractive effect. These effects are associated to the optical damage which is controlled for holographic applications but is a drawback for telecom ones. Various techniques exist for measuring these effects and among them, we present quite direct measurements techniques useful for qualification of materials. Within these techniques, various parameters can be taken into consideration as the temperature, the wavelength and the power of the laser beam and additional physical properties of the material under study can be determined as the activation energy or the damage threshold. Some results obtained with pure and doped lithium niobate crystals, illustrate this study.

Key words: Lithium niobate, electrical conductivity, energy of activation, photo-conductivity, photo-refractive, damage, threshold

INTRODUCTION

Before integration of material in devices, a large phase of qualifications is necessary to determine its functional and physical properties in the aim of a qualification and an enhancement of its efficiency. This experimental phase has to take into account, the global set of properties and parameters involved in the practical process. In optoelectronic devices, the functional properties are linked to the wavelength and power of lasers used in the application.

A first example is in optical modulators where a single laser is used in the infrared telecom range at a few milliwatts. A second example is in all-optical laser amplifier in which a pump-probe arrangement involved two wavelengths at hugely different powers. Finally, we can also site holographic applications in which two beams are necessary for writing and reading the dynamic structure. A common point that we can notice in these examples is the importance of the photo-refractive effect which is the effect describing the influence of the optical beam on the refractive indices.

Indeed, this effect is mainly influence by the conduction effect and it determines the resistance to the optical damage of the material that we may want high or low depending of the type of applications, e.g., telecom or holographic applications, respectively. In this contribution, we present measurement techniques of two fundamental properties in the field of optoelectronic that are the electric or dark conductivity and the photo-conductivity, allowing the determination of the optical damage. We have selected techniques that permit to take into account, external physical parameters such as the temperature and the laser power used for the experiment and to reach back to the determination of some physical parameters of materials under study such as the activation energy or the photo-refractive specific time.

Finally, we illustrate the experimental techniques with results obtained in the characterization of a set of lithium niobate, LN crystals. The choice of this sample family is driven by the fact that lithum niobate is a key material in optoelectronic due to its huge versatility.

EXPERIMENTAL TECHNIQUES

The phenomena of charge transport in LN without and under illumination in usual conditions, i.e., visible or near infrared light at room temperature up to far the phase transition temperature (Tc = 1200°C), imply only the electrons of conduction.

In the following, we present and comment techniques for the measurements of dark and photo-conductivities with numerous insulating materials, i.e., with crystals presenting weak electrical conductivity and we discuss the experimental conditions necessary to follow for obtaining a high accuracy in the measurements.

Corresponding Author: B. Chikh-Bled, MOPS Laboratory, Paul Verlaine University of Metz and Supelec, 2 rue Edouard Belin, 57070 Metz, France

Dark conductivity measurement technique: Electric conductivity, named dark conductivity in optoelectronic is one of the most delicate properties to measure in insulating materials like lithium niobate. Measurements of weak currents can be prone to many sources of errors degrading the precision of measurements. All techniques require sample of great surface and low thickness as well as the deposit of particular electrodes. In a material of very significant voluminal resistance placed between two electrodes to which one applies a potential difference, the measured total current will be the sum of the current crossing the volume of the sample and that crossing the surface of material.

To determine the volume conductivity, it is necessary to be able to free itself from the surface conductivity. For this, we used the technique of guard which consists in depositing on a face of the crystal the central electrode largest as possible, giving the maximum possible current surrounded by an electrode referred as the guard electrode and on the other face, a central electrode in a straight forward opposite position than this on the 1st face (Fig. 1). It results from this that the surface current will be equal to zero when considering the resistance of the surface as infinite. During experiment, the electrode of guard is connected to the same potential as the central electrode. Thus, one measures only the electrical current crossing the volume of the material. On the two faces of the crystal, we entirely stuck scotch tape with ugly of the laser cutting machine, we cut out the yellow squares (Fig. 1).

In the preparation phase of experiments, we have masked each face following the above schema or equivalent shown in Fig. 1. We have sputtered gold electrodes by an evaporation technique. For measurements, the crystal is placed on a ceramic plate having the advantage of a high temperature reliability and a high electrical resistance. In contrary, it is well known that its main disadvantage is the difficulty for its machining. To bring thermal energy to the crystal, the unit carries sample-crystal is placed in a furnace.

In Fig. 2, we present the experimental setup used for dark-conductivity characterisations of LN crystals in the laboratory. This setup is based on a Source Measure Unit (SMU) designed for current measurements in the nA to the μA ranges. A voltage source is connected at the boundaries of the crystal. Finally, a thermocouple of K type (chromium-alumel) controls the temperature of the crystal. This type of sensor is well known for its robustess and for its very broad temperatures range in which it can be used (it tolerates temperatures of several hundreds of degrees, it makes it possible to measure between 10 and 570°C).

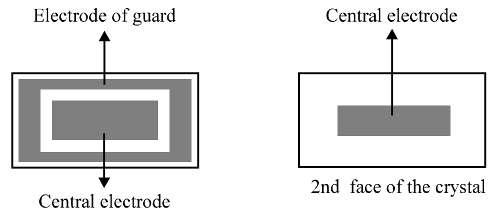

Fig. 1: Realization of the electrodes on the two faces of the crystal

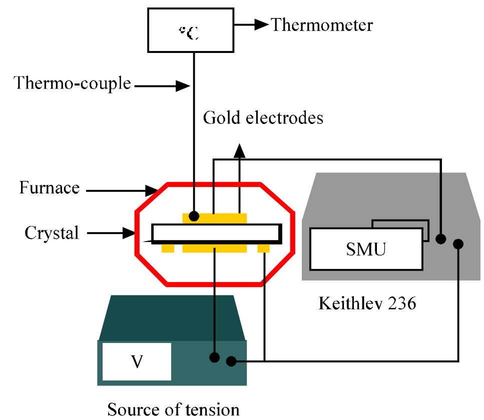

Fig. 2: Experimental setup for electric-dark conductivity measurementss

Photo-conductivity and optical damage measurement technique: The optical damage in LN and crystals of its family is mainly due to the charges (electrons in these crystals) displacement induced by an illumination. In these crystals, the photoinduced charges displacement induced, via the electro-optic effect, the variation of the refractive indices. It is the reason why the optical damage is also named in this case photo-refractive damage (Kostritskii *et al*., 2010). This phenomenon is governed by the photo-conductivity depending of the wavelength and the power of the beam. To investigate the kinetics of the photo-refractive response in LN crystals, we have implemented the closed aperture pseudo-Z-scan arrangement.

Within this setup, the associated method consists of the recording in the far field region throughout a pinhole of the dynamic optical response, i.e., the photo induced variation of the output laser beam of a sample placed at a focal point (Kostritskii *et al*., 2010). In Fig. 3, researchers shows the experimental setup used for photo-conductivity characterisations of LN crystals in the laboratory.

A CW beam emits by a laser diode at 644 nm with an emitted power equal to 65 mW is focused at the entry of the sample S with a 200 mm focal Lens (L). The direction of the input beam Polarization (P) is adjusted by

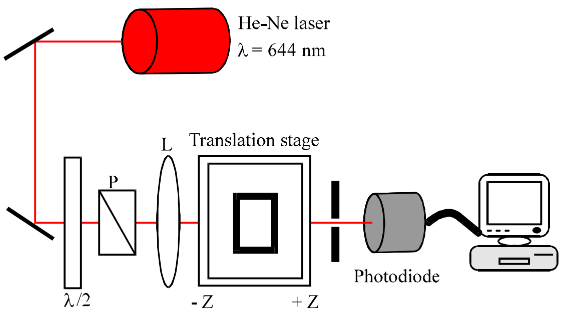

Fig. 3: Experimental setup based on the pseudo Z-scan arrangement used for studying the optical damage

rotating a half-wave plate (λ/2). A translation stage enables to vary with precision, around the focal point of the lens (z = 0), the position Z of the crystal maintain on a goniometric head. At 300 mm after the focal point of the beam, i.e., in the far field region, a Diaphragm (D) is adjusted in the initial phase of the experiment, i.e., without sample to insure a half of the power of the beam which was detected by a silicon photodiode with an active surface of 15 mm^2. A computer controls the translation stage and the data acquisition with a program developed under Lab view.

Application to lithium niobate crystals: The methods described previous were successfully applied to a set of lithium niobate crystals. To illustrate this study, we show significant results obtained in pure and iron doped LN crystals.

Thus, we present experimental results obtained with the dark-conductivity measurement technique in a congruent pure LN crystal which have a weaker electrical-conductivity than the iron doped crystals. Complementarily, researchers present results obtained with the photo-conductivity and optical damage measurement technique in iron doped LN samples which have shorter illumination response time than pure crystal.

Dark conductivity of pure lithium niobate crystals: The congruent pure LN crystal used for this experiment is a x-cut wafer of 700 micrometers width and x faces allowing 2 cm^2 central electrodes. Figure 4 shows the current recording by the SMU when voltage is applied onto the crystal electrodes. To point out the main information given by the experimental data, the response for voltage up to 200 V even if they were done up to 400 V. These measurements were performed in the 160-300°C temperature interval. Under 160°C, no current was detected because the thermal contribution to the energy for the voltage applied to the crystal is insufficient to excite electrons towards the conduction band and only

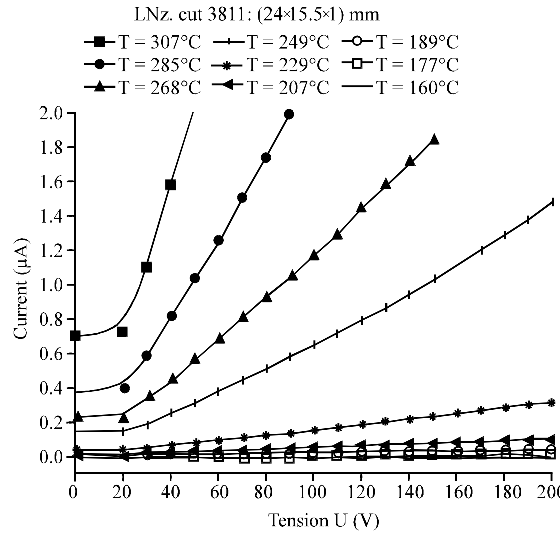

Fig. 4: Experimental current and tension values for various temperatures

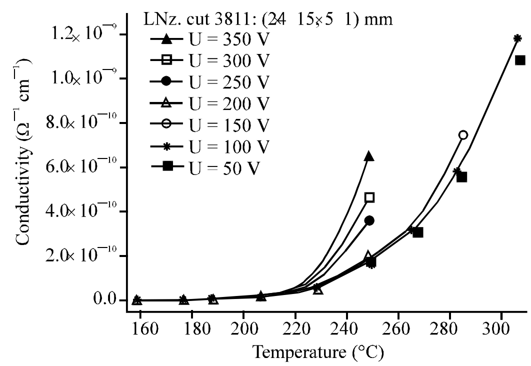

Fig. 5: Evolution of electric conductivity with the temperature for the various tensions applied

above 160°C, currents have been measured. It is to be of note that between 160 and 207°C, the behaviour for each temperature of the current according to the applied voltage is quasi-linear. On the other hand, starting from 229°C, one notes a beginning of non non-linearity. In the 200-300°C range, the voltage threshold decreases when the temperature increases resulting in a conduction mechanism in which the charge carriers are released by the thermal energy action. We notes that the temperature necessary for a thermic action of the electrons sufficient to obtain conduction is located around 230°C. From these experimental results, we have shown in Fig. 5, the conductivity as a function of the temperature for the whole voltage range. To extend the main goal of this contribution devoted to experimental aspects of the conductivity measurements, we briefly explain the behaviours shown in Fig. 5 and for that we remember that

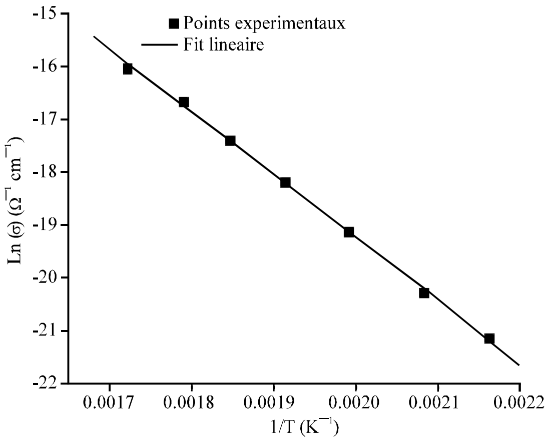

Fig. 6: Electric conductivity in the interval of temperature 189-300°C

LN being a strongly resistive material with its resistance that decreases when the thermal contribution of energy increases; this phenomenon results in an increase of the electrical current and consequently an increase in conductivity. The dependence in temperature of the electric conductivity in interval 189-300°C is obtained by tracing the logarithm of the conductivity according to the inverse of the temperature from the values of resistances obtained for each temperature. This layout is shown in Fig. 6 and point out that the electric conductivity obeys the Arrhénius law (Kostritskii *et al*., 2010) described by Eq. 1:

$$\sigma = \sigma_0 \exp\left(-\frac{E_a}{K_B T}\right) \quad (1)$$

Where, E_a, σ_0 and T are the activation energy, the pre-exponential factor at a given temperature and the absolute temperature of the material, respectively, K_B is the Boltzmann constant. The slope of the graph above enables us to determine the activation energy, found equal to E_a = 1.031 eV. By extrapolation, the electric conductivity at 600°K of the pure congruent LN sample is found equal to $\sigma = 2.29\times10^{-9}\ \Omega^{-1}\ cm^{-1}$ in quite good agreement with literature (Mansingh and Dhar, 1985) when we take into account, the variations of conductivity linked to the possible differences of compositions and impurities presents in the crystal. Indeed, Klauer *et al*. (1992) show that the activation energy varies from 0.97 ($\sigma = 8.59\times10^{-9}\ \Omega^{-1}\ cm^{-1}$) to 1.17 eV ($\sigma = 1.8\times10^{-10}\ \Omega^{-1}\ cm^{-1}$) for a crystal of LN containing of iron (Yang *et al*., 2001) therefore, we note that the computed value of E_a for the crystal (Z cut 3811) is well with this interval (Klauer *et al*., 1992).

Photo-conductivity and optical damage of iron doped lithium niobate crystals: The crystals used for this experiment are two congruent LN crystals LN3AG and LN5AG, doped with 0.03 and 0.05% at. of iron, respectively. These crystals are considered as As-grown crystals (AG) as they did not received any additional post growth treatments as oxidation or reduction ones. Their dimensions are (x, y, z) = (5,7,10) mm^3 and placed on a sample holder with for the laser beam at 644 nm, a propagation axis parallel to x and a polarization along the z axis of the crystals. The shape and the waist size of the laser beam are measured by the Knife edge method at several focal distances. At the focal position, the beam presents a waist equal to 0.07 mm. The beam power control is obtained by insertion of ten density filters allowing a power density varying from 0.4-100 W cm^{-2} at the focal point.

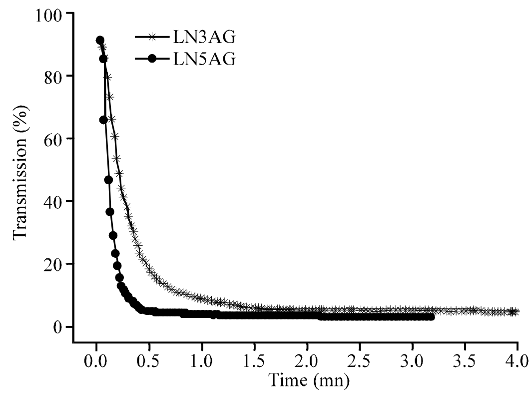

Fig. 7: Experimental optical damage kinetics in iron-doped samples at input power density equal to 42 W cm^{-2}

Under these experimental conditions for the two crystals, the dynamic dependences of the intensity of the beam at a power density equal to 42 W cm^{-2} at the focal point are showed in Fig. 7.

The curves in Fig. 7 clearly show the developing of the optical damage usual in these iron doped LNl. All the curves were adjusted at an initial value of the transmitted beam intensity equal to 100% to take into account, the absorption of the samples that is largely different between both at the laser wavelength, 644 nm used in these study. During a first qualitative analysis of these experimental results, we can see that the response time of the As-grown crystals, LNAG depends directly on the absolute iron concentration and that the crystals having the strongest iron concentration have a shorter response time.

We can note also that the absorption is not responsible for the damage because both crystals present the same damage threshold at long times. The photo-conductivity was calculated using Eq. 2 from the experimental results of the photo-induced distortion of the

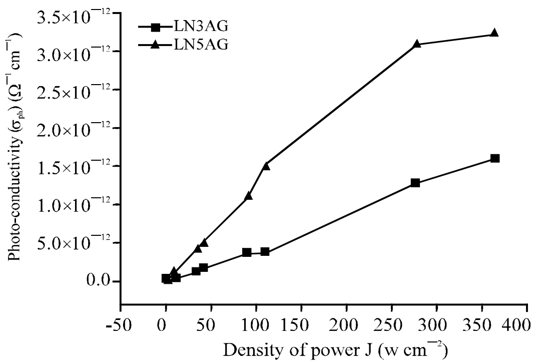

Fig. 8: Photo-conductivity as function of the input power density in iron-doped samples

transmitted light spot (Peithmann *et al*., 2002) as showed in Fig. 3 for the power density varying from 0.4-100 W cm^{-2} at the focal point:

$$\sigma_{ph} = \frac{\varepsilon \varepsilon_0}{\tau} \quad (2)$$

Where:

ε = 30 is the dielectric constant of LN

ε_0 = Permittivity of free space (ε_0 = 8.854×10^{-12}) (Kostritskii *et al*., 2010)

τ = Characteristic relaxation time

The variation of the photo-conductivity of the two samples LN3AG and LN5AG according to the power density of illumination is showed in Fig. 8. For the two samples, the curves showed in Fig. 8 point out a quasi-linear dependence of the photo-conductivity according to the power density of the illuminated beam in the operated range. Finally, a comparison between the two curves shows an increase of the photo-conductivity according to the iron concentration.

CONCLUSION

With the goal of optical damage characterisation of crystals that have to be integrated in opto-electronic devices, we have presented, analysed and discussed two experimental methods with their dedicated arrangements for the determination of the dark and photo-conductivities which are the two main physical processes involved in photorefractive materials. We have showed that with these methods can take into consideration various parameters as the temperature, the wavelength and the power of the laser beam.

From the analysis of the experimental, results obtained within these techniques, additional physical properties of the material under study can be determined as the activation energy or the damage threshold. To justify these proposals, we applied the two experimental methods for the characterisation of samples taken in the lithium niobate family which presents the family mostly used in opto-electronic applications as substrats or bulk crystals. When some experimental precautions are taken, the two techniques proposed even with their quite simplicity, give reliable results in the determination of the dark and photo-luminescences.

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