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Proton exchanged depth determination in $LiNbO_3$ from laser induced pyroelectric voltage measurements

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We report a new nondestructive technique for the determination of depth of proton exchanged (PE) layers in $LiNbO_3$. Relying on the difference in the magnitudes of measured pyroelectric voltages, induced by an intensity modulated laser beam, on the exchanged and unexchanged regions, the depth of the PE layer has been measured and corroborated by a conventional optical characterization method.

Proton exchange (PE) process has become an important technique for the fabrication of optical and acoustical waveguides.[1] $LiNbO_3$ and $LiTaO_3$ are two materials in which the PE process and the resultant PE substrates have been extensively modeled and characterized.[2–5] PE has generally been achieved using benzoic acid or a mixture of benzoic acid and lithium benzoate as the source, but pyrophosphoric acid has also been used for this purpose.

The depth of the exchanged region is a critical parameter of the waveguide, determining the number of propagating modes. It is generally estimated from relatively involved techniques such as inverse Wentzel–Kramer Brillouin (IWKB) optical characterization,[6] Rutherford backscattering spectrometry (RBS), and nuclear reactions,[2] etc., which are either complex, expensive, and/or destructive.

Recently, we reported a new, nondestructive evaluation (NDE) method of experimentally determining the exchange depth by means of phase measurements of laser induced pyroelectric signals.[7] Subsequently, we found that the same information can be independently derived from the pyroelectric signal magnitudes also. Its analysis and experimental verification is presented below in this letter. The basis of this NDE technique is the pyroelectricity of the substrate material, e.g., $LiNbO_3$ and $LiTaO_3$. The process of PE forms a thin insulating overlay on these substrates. An intensity modulated laser beam, incident on the surface, causes modulated heating which induces a pyroelectric voltage at the interface between the PE overlay and the underlying substrate.

Imaging based on pyroelectric detection is a special case of thermal wave imaging. Modulated heating of a given solid surface results in the propagation of a thermal wave,[8] the wavelength, λ, of which is given by

$$\lambda=(4\pi K/f\rho c)^{1/2}, \tag{1}$$

where K, ρ, and c are, respectively, the thermal conductivity, mass density, and specific heat of the material, and f is the modulation frequency of the radiation source (optical, e beam, etc.). In general, the problem of unidimensional conduction of heat in a solid undergoing periodic surface heating can be described mathematically by the differential equation,[8]

$$\frac{(\delta^2 T)}{(\delta z^2)}-\left(\frac{c}{K}\right)\frac{(\delta T)}{(\delta t)}=0, \tag{2}$$

where T is the temperature assumed varying in the z direction. The temperature modulation of thermal wave has a solution of the form

$$T(z,f,t)=Ae^{-2\pi z/\lambda}\cos 2\pi(ft-z/\lambda). \tag{3}$$

The method of pyroelectric imaging, utilized here, converts the temperature modulation directly into an electrical signal. Whenever a pyroelectric material undergoes a change of temperature (ΔT), surface charge (ΔP) is produced as a result of the change in its spontaneous polarization:

$$\Delta P=p\Delta T, \tag{4}$$

where p is the pyroelectric coefficient. Therefore, a propagating thermal wave, encountering a pyroelectric material in its path, would generate a surface charge. The modulated heating modulates the surface charge and the resultant alternating pyroelectric current/voltage can be electrically measured through metal electrodes deposited across the sample faces.

A typical sample cross section (Fig. 1) shows a pyroelectric substrate exchanged in part of its top surface, with both top and bottom faces metallized to form electrodes. The heating modulation frequency is chosen such that the corresponding thermal wavelength $\lambda \ll d$, the sample thickness. When an intensity modulated laser beam is incident at a point on the non-PE region of the sample, the voltage

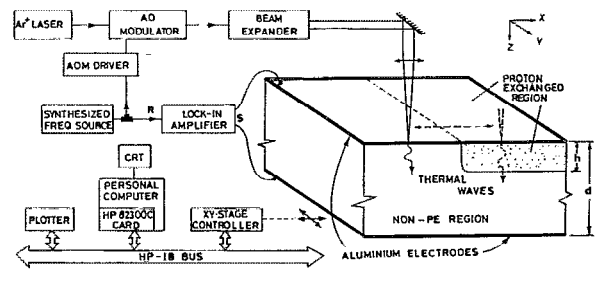

FIG. 1. Schematic configuration of the automated setup for pyroelectric measurements, showing a typical sample cross section for illustrating the technique of PE depth measurement from magnitude information of pyroelectric signals.

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developed and measured (magnitude/phase) is, say, V/ϕ. Similarly, if the same beam is now made incident at a point on the PE region, a different voltage, say, V_{PE}/ϕ_{PE} is measured. This suggests that the process of proton exchange causes a change in the pyroelectric constitution of the sample. Reports that in the process of PE, 70% of Li atoms are exchanged by protons,[2] and also that the PE layer is virtually nonpiezoelectric,[3] reasonably validates the assumption that the PE layer is rendered nonpyroelectric as well. We also assume that for a given modulation frequency f, the thermal wavelength λ is the same in both the PE and unexchanged regions. Thus, for a first order modeling the PE layer is assumed nonpyroelectric but otherwise "thermally" unchanged. Let h be the PE layer depth to be determined from measurements of the pyroelectric signal generated at points in the virgin and exchanged regions. In the former, the signal is generated between the two, surface, metal electrodes. In the latter case, in the PE region, the pyroelectric signal is generated only after the thermal wave, generated on the surface, has traversed a distance h, and is then incident at the layer-substrate interface.

We now analyze the observed change in the magnitudes of the pyroelectric voltages in the PE and unexchanged regions. For a given modulating frequency, Eq. (3) gives the decay with depth of the amplitude of the temperature oscillation. Equation (4) relates the polarization of the pyroelectric to its temperature. In the case when the laser beam is incident on the PE region, the pyroelectric signal is generated when the thermal wave generated on the surface reaches the PE layer-substrate interface. Having traversed a distance h, the decrease in amplitude of the temperature due to thermal wave oscillation and thus the resulting pyroelectric voltage can be calculated. Knowing this voltage, the PE depth can be calculated as

$$h=(\lambda/2\pi)\ln(V/V_{PE}). \tag{5}$$

A Z-cut $LiNbO_3$ wafer, 1.5 cm$\times$1.5 cm$\times$390 μm, with one face optically polished and the other finely lapped, was proton exchanged on part of its polished surface. A thick aluminum film was used as a mask for the unexchanged region, when the sample was PE at 240 °C in benzoic acid. Measurements were made using a Hewlett Packard Interface Bus (HPIB) interface automated experimental setup[7,9] for laser induced pyroelectric signal generation, acquisition, and display, which had been utilized earlier for obtaining the first ever reported pyroelectric imaging scans of PE $LiNbO_3$.[9] It is an improved version of the setup employed for pyroelectric NDE of bulk acoustic wave transducers.[10] A focused, acousto-optically intensity modulated Ar^+ laser beam is made normally incident on the sample surface (Fig. 1). By movement of the automated X-Y scan stages (minimum step size of 1 μm), on which the sample is mounted, the laser beam can be made incident on the PE and unexchanged regions. The pyroelectric voltage thus developed is measured, across a 0.5 MΩ resistor in parallel with the sample, by an interfaced lock-in amplifier.

At a modulation frequency of 2.21 kHz, corresponding to a thermal wavelength of 89.4 μm in $LiNbO_3$, the voltage magnitudes measured were $V=2.76$ μV and $V_{PE}=2.37$ μV. This, from Eq. (5), gives a PE layer depth of 2.17 μm. This was corroborated by measurements at other frequencies, i.e., at 6.67 kHz the measured voltages ($V=1.08$ μV) and ($V_{PE}=0.83$ μV) indicate a PE depth of 2.15 μm.

These compare very favorably with the depth of 2.2 μm, determined from the inverse WKB technique[6] using effective indices measured by the conventional prism coupling method.

An interesting future extension would be the derivation of the PE layer depth from a measurement in the exchanged area only, without reference to the virgin region, e.g., for planar waveguides. This seems possible only from the amplitude of the pyroelectric signals (being absolute measurements), and not from the phase which is relative. In this respect, it may be mentioned that our measurements of the (absolute) pyroelectric voltages are in conformity with the reported pyroelectric models.[10] The pyroelectric voltages measured are developed due to the flow of pyroelectric current across the fixed value parallel resistor and are therefore directly proportional to the short-circuit intensity i_{cc} of the pyroelectric current given by

$$i_{cc}=-Q_a({}^{PC}p_z/d\rho c), \tag{6}$$

where ${}^{PC}p_z$ is the planar clamped pyroelectric coefficient for the z axis perpendicular to the sample surface and Q_a is the power which diffuses into the substrate of thickness d. Physically, it is this diffused power which gives rise to a temperature modulation and its variation is thus given by an expression similar to Eq. (3) for the temperature. When the modulated laser is on the unexchanged region, the pyroelectric current and voltage correspond to the diffused power Q_0 at the surface $z=0$. The voltage developed, when the same laser beam is now on the PE region, is then due to a decayed power Q_{PE} at $z=h$

$$Q_{PE}=Q_0\exp(-2\pi h/\lambda). \tag{7}$$

At the heating frequency $f=6.67$ kHz, $V=2.76$ μV, $i_{cc}=5.52$ pA, and Eq. (6) gives $Q_0=83.89$ μW. In the PE region, $V=2.37$ μV corresponds to a $Q_{PE}=72.04$ μW. Therefore from Eq. (7), the PE layer depth of 2.17 μm is, likewise, in excellent agreement with previous results.

In conclusion, a new nondestructive technique for measuring PE layer thickness in $LiNbO_3$ crystals using pyroelectric signals, extendable in principle for diffused/implanted/exchanged layers in all pyroelectric materials as well, is presented. The immediate relevance to optical and acoustic waveguide substrates like $LiNbO_3$ and $LiTaO_3$, by providing a NDE alternative to RBS and secondary ion mass spectroscopy (SIMS), as well as rivaling prevalent optical characterization schemes, can be enhanced by future extensions of the technique, e.g., for analyzing stripe waveguides the incident modulated laser beam should be finely focused to a few micrometers. Interesting possibilities also exist in deriving thermal and other material properties of the PE layers/substrates.

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