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Holographic Measurement of Dark Conductivity in LiNbO₃:Ti:Fe Planar Optical Waveguides¹)

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LiNbO₃ is a promising photorefractive material for many applications [1]. Phase holograms formed in strip and planar waveguides may be used for phase conjugation, narrow-band holographic filters, or light amplifiers [2]. In most cases $Fe^{2+/3+}$ serves as the photorefractive center. On the one hand, the maximum value of the light-induced refractive index changes has been found to be proportional to the concentration of Fe^{3+} [3]. On the other hand, there are several hints that iron doping also has a significant influence on the dark conductivity of the material [4], thereby limiting the storage time. In waveguides the Fe concentration can be easily varied by indiffusion. In this letter we will show that the dark conductivity of heavily iron-doped planar waveguides in LiNbO₃ increases exponentially with the iron concentration, and this correlation may also be applicable to samples with a much lower doping.

Planar photorefractive waveguides are fabricated by two subsequent surface diffusions using commercially available optical grade y-cut LiNbO₃ wafers of congruently melting composition. At first, an 80 nm-thick evaporated titanium layer is indiffused at 1000 °C for 24 h in wet argon atmosphere to increase the surface refractive indices of LiNbO₃. In a second step, evaporated iron layers of different thicknesses (30, 40, 60, 80, 100, and 120 nm) are indiffused at 1000 °C for 18 h in wet oxygen to increase the photorefractive sensitivity of the samples. For comparison, we investigate a titanium-diffused waveguide (80 nm-thick Ti layer, indiffused at 1000 °C for 18 h in wet oxygen) on a homogeneously doped LiNbO₃: Fe substrate [5], too.

The concentration profiles of both, titanium and iron, can be well described by Gaussian functions, $c_i = c_i^0 \exp(-y^2/2q^2)$, with penetration depths $q_{Ti} = 3.6 \,\mu\text{m}$ and $q_{Fe} = 10.7 \,\mu\text{m}$, respectively [6]. All waveguides are multimode with typical depths of the nearly Gaussian refractive index profiles of $q_{wg} \approx 5 \,\mu\text{m}$. The relative iron concentration at the surface is measured with the help of an electron microprobe, where we demonstrate that c_{Fe} is proportional to the thickness of the evaporated iron layer. The absolute value of the surface concentration of the indiffused iron, $c_{Fe}(y = 0)$, is determined by X-ray photoelectron spectroscopy (XPS) [6]. Only Fe³⁺ can be detected, while the concentration of Fe²⁺ is below the measuring accuracy. Thus we estimate a concentration ratio $c_{Fe^{2+}}/c_{Fe^{3+}}$ well below 0.01.

The dark conductivity in the waveguide is measured with a holographic two-wave mixing technique. Both, a TE and a TM mode are excited with the help of a rutile prism to propagate collinearly along the x-axis of the sample, and the transmitted powers are measured. Modulated photovoltaic currents are generated by these orthogonally polarized waves, and a periodic space charge field builds up which leads to a perturbation of the dielectric constant via the electrooptic effect. Details of the formation of the gratings can be found in [7,8]. When the phase gratings are written to saturation, the two writing beams are switched off, and the decay of the grating in the dark is monitored by subsequently measuring the diffraction efficiency for short time intervalls (50 ms) with a low-intense reading beam. We define the diffraction efficiency as the ratio of diffracted to total read-out light intensity. The dark conductivity σ_d is calculated from the relaxation time τ_d of the gratings via $\sigma_d = \varepsilon \varepsilon_0 / \tau_d$, where ε_0 is the vacuum permittivity and ε is the static dielectric constant of LiNbO₃.

In Fig. 1 the measured value of dark conductivity is shown as a function of the iron concentration $c_{\text{Fe}} \approx c_{\text{Fe}^{3+}}$ at the surface of different waveguides. Samples with iron contents larger than $2 \times 10^{20} \text{ cm}^{-3}$ are produced by indiffusion of iron, whereas the sample with the lowest iron concentration ($c_{\text{Fe}} = 0.13 \times 10^{20} \text{ cm}^{-3}$) is fabricated on a LiNbO₃:Fe substrate that has been homogeneously doped in the melt. For the different modes of a waveguide, i.e., for different propagation depths, we obtain almost the same dark conductivity. This is expected from the nearly constant iron concentration in the waveguiding layer, and points to a minor influence of titanium on dark conductivity, because the titanium concentration strongly decreases with increasing propagation depth [6].

The measured dependence $\ln [\sigma(c_{\text{Fe}})]$ can be fairly well approximated by a straight line, including the sample prepared on the melt-doped substrate, thus obeing the relation $\sigma_d(c_{\text{Fe}}) = \sigma_d^0 \exp (c_{\text{Fe}}/c_{\text{Fe}}^0)$,

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Fig. 1. Dark conductivity σ_d as a function of the surface iron concentration $c_{Fe}(y=0)$ of different LiNbO₃:Ti:Fe planar waveguides. Samples with iron concentrations larger than 2×10^{20} cm⁻³ are fabricated by indiffusion of iron, whereas the sample with the lowest iron concentration ($c_{Fe} = 0.13 \times 10^{20}$ cm⁻³) is fabricated on a LiNbO₃:Fe substrate that has been homogeneously doped in the melt

with the parameters $\sigma_d^0 = 8.3 \times 10^{-17} \ (\Omega \ cm)^{-1}$ and $c_{Fe}^0 = 0.87 \times 10^{20} \ cm^{-3}$. This behavior may be explained by hopping conduction, where delocalized, excited electron states of neighbouring Fe²⁺ centers (or other defects that are connected with iron) overlap with each other. For this type of dark conduction an exponential dependence on impurity concentration is predicted [9]. This points to a nearly constant concentration ratio $c_{Fe^{2+}}/c_{Fe^{3+}}$ in our strongly oxidized samples, as it is expected from the identical fabrication conditions and diffusion atmospheres.

In the same way as described above, titanium-diffused waveguides have been fabricated that are additionally doped with copper instead of iron. As a first result, in these samples, even for high doping levels, much smaller dark conductivities have been observed when compared with the iron-doped samples. Consequently, for applications where both, high light-induced refractive index changes and low dark conductivity are required, copper doping instead of iron may probably be advantageous.

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