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OPTICAL WAVEGUIDES IN LiTaO_3 *V. V. Atuchin and C. C. Ziling**Institute of Semiconductor Physics, Siberian Branch of the USSR
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Abstract. The optical properties of waveguide layers produced by Ti, Zr or Hf diffusion into LiTaO_3 have been studied. A model of mass transport in Ti:LiTaO₃ system taking into account the prolonged existence of Ti source on waveguide surface has been proposed and confirmed by experiment. A method for controlling the Li/Ta ratio in LiTaO_3 crystals by birefringence measuring has been developed. The dependence of Ti:LiNbO₃ and Ti:LiTaO₃ waveguide characteristics on crystal stoichiometry has been found and explained.

Резюме. Определены оптические характеристики волноводных слоев, полученных диффузией Ti, Zr, Hf в LiTaO_3 . Предложена и экспериментально подтверждена модель массопереноса в системе Ti:LiTaO₃, учитывающая длительное существование эффективного источника титана на поверхности волновода. Предложен метод контроля состава подложек LiTaO_3 по двулучепреломлению. Обнаружена и интерпретирована зависимость параметров волноводов типа Ti:LiTaO₃ и Ti:LiNbO₃ от состава подложки.

Lithium niobate (LN) is now one of the most widely used material in integrated optics because of its favourable electrooptic properties and commercial availability. However, the photorefractive index changes in Ti:LiNbO₃ waveguides give rise to a long time drift of device parameters in particular at visible and near IR wavelengths [1, 2].

On the other hand, since LiTaO_3 (LT) resists "optical damage" by about two orders of magnitude as much as that of LN, it seems to be a more attractive material for optical devices.

From the practical point of view the waveguides fabricated by Ti-diffusion in LT are interesting because of their low level of losses and high index increase (~ 0.02) in the visible region of spectrum. Although this method of waveguide formation has been available for more than a decade [3], the principal parameters of such layers are still unknown.

The mode dispersion in diffused planar waveguides is defined completely by the profile of the diffusing substance and the dependence of indices increase on the admixture concentration. With the aim of choosing the diffusion model, that is able to describe the real process of mass transport, and finding the correlations between the Ti concentration and the index rise, the effects of fabrication conditions on the refractive index profile of planar Ti:LT waveguides has been studied. As demonstrated in [4], the increase of extraordinary Δn_e and ordinary Δn_o index of LT is a linear function of the Ti concentration. If the diffusion time is long enough, the profiles of Δn and

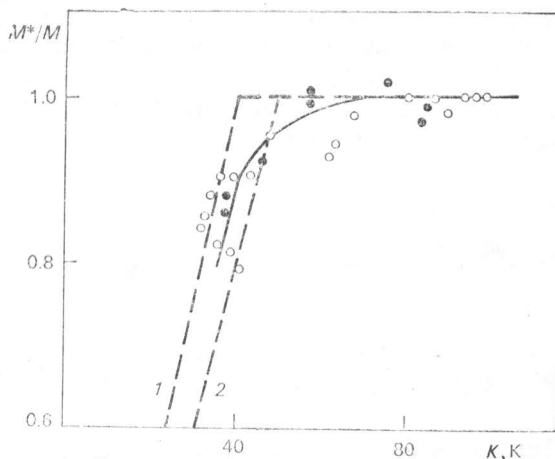
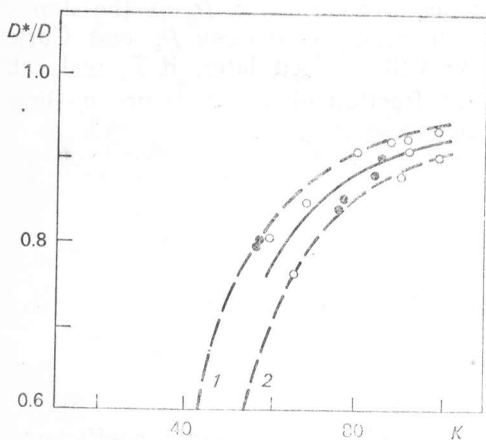
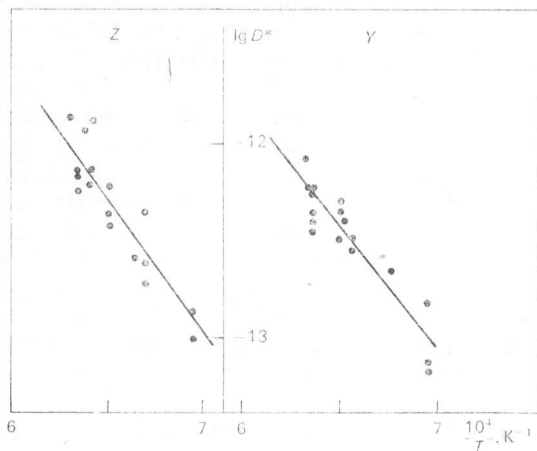


Fig. 1. The dependence of Ti-diffusivity D^* in Y and Z-cuts of LT on temperature
 Fig. 2. The dependence of M^*/M on K : \circ — Z-cut; \bullet — Y-cut; 1) $K^*=40$; $K^*=50$
 Fig. 3. The dependence of D^*/D on K : \circ — Z-cut; \bullet — Y-cut; 1) $K^*=40$; 2) $K^*=50$

Ti concentration can be approximated by a Gaussian function. The titanium diffusivities D^* in Y- and Z-cuts as a function of temperature are shown in Fig. 1. One can see that noticeable anisotropy of D^* is absent. The activation energy of the process is equal to 2.6 eV, that is near the corresponding value for the Ti:LN system.

In the initial stage of the reaction a compound is formed from the TiO_2 and various components in LT and further this surface film acts as a continuous source of Ti, that results in deflexion from the model of diffusion from the instant source. Figures 2 and 3 show the dependences of the indiffused fraction of titanium M^* and D^* on $K=\theta/h$, where θ is a diffusion length and h is an initial thickness of Ti. There are functions calculated on the base of the diffusion model, that take into account the prolonged existence of Ti-source on the waveguide surface [5]. Using the expression derived in [5] for the dependences of M^*/M , where $M=\rho$, ρ is Ti density, and D^*/D^* , where D is a real diffusion coefficient, on h , one can estimate the applicability of the instant source model for the discription of Ti diffusion in LT. The condition $M^*/M \geq 0.9$ is

Table 1

Cut	Y	Z
$T^{\circ}\text{C}$	1300	1300
K	45	45
c_1^* , kg/m ³	180	180
c^* , kg/m ³	200	200
$D \times 10^{11}$, m ² /S	5.3 ± 0.3	7.6 ± 0.3

satisfied, if $K \geq K^* = \sqrt{\pi} \rho / c^*$, where c^* is the upper limit of solid solubility of an admixture. The condition $D^*/D \geq 0.9$ is satisfied if $K \geq 2K^*$. The parameters of Ti diffusion in LT defined with allowance for the above mentioned limitations are shown in Table 1. As the upper limit of Ti solubility in the waveguide layer c_1^* and the corresponding quantity c^* in $\text{TiO}_2\text{:LT}$ ceramic are similar, it can be supposed that the decrease of Ti content during surface

doping corresponds to the transference nearly in parallel with the composition line $\text{TiO}_2\text{-LT}$ considered in the ternary diagram $\text{Li}_2\text{O-Ta}_2\text{O}_5\text{-(TiO}_2)_2$.

Physical mechanism causing the refractive indices variation in doped layers of lithium niobate and tantalate are of considerable interest for the fabrication of a wide variety of a waveguide structures and, in particular, for the conscientious search of new admixtures.

According to [6], there are three possible mechanism for the waveguide formation in ferroelectrics, are follows: due to increased electronic polarizability (or molecular refraction) R by doping, due to decreased spontaneous polarization P_s of the doped crystal, and due to the photoelastic effect. The relation [7] between P_s and Curie temperature T : $T \approx BP_s^2$, where $B \approx 3 \times 10^3 \text{ deg. m}^4/\text{C}$, will be used later. If T_c and cell parameters vary in direct proposition to the molar fraction of an admixture y , then, taking into account the condition $\Delta n \ll n$, we can write [8]:

$$\Delta n_{e,0} = A_{e,0} y$$

$$A_e = \frac{(n_0^2 + 2)^2}{6n_0V} \frac{\Delta R}{y} - \frac{n_e^3}{2B} g_{33} \frac{\Delta T_c}{y} - \frac{n_e^3}{2} p_{31} \frac{\Omega}{y},$$

$$A_0 = \frac{(n_0^2 + 2)^2}{6n_0V} \frac{\Delta R}{y} - \frac{n_0^3}{2B} g_{13} \frac{\Delta T_c}{y} - \frac{n_0^3}{2} p_{11} \frac{\Omega}{y},$$

where V is the molar volume of LiTaO_3 , g_{ij} are the quadratic electrooptic coefficients, v_{ij} are the photoelastic coefficients, and Ω is the quantity corresponding to elastic deformation [9].

To calculate the refraction term, it is expedient to use the molecular refraction of suitable oxides since the ion refraction of the elements is strongly dependent on the ionicity of chemical bounds. The variation of T_c and cell parameters as a function of y can be obtained from the ceramic data. The calculated values $A_{e,0}$ and those founded by experiment for a few LT-based solid solutions are shown in Table 2. The experimental values of $A_{e,0}$ are in the last column. All data are for wavelength equal to 632.8 nm.

One can see that the concurrence of theoretical and measured values of A may be qualified as semiquantitative.

The first line of Table 2 collects the separate terms and the summary values of $A_{e,0}$ calculated for the stoichiometry deviation of LT indices. Figure 4 shows the variation of the LT indices and birefringence with decreasing lithium content. To analyse the magnitude of every term, it is evident that the n_e should increase mainly induces by the p_s decrease with Li depletion, but the n_0 constancy takes place as a consequence of the compensation between all the three increments.

Table 2

	A	(R)	(P _s)	(ε)	Σ	Σ _{exp.}
Ta: LiTaO ₃	A _e	(-0.057)—(-0.040)	0.339	-0.002	0.280—0.297	0.256 ± 0.024
Li _{1-y} Ta _{1+y/5} O ₃	A _o	(-0.057)—(-0.040)	0.086	0.001—(0.015)	0.114—0.047	0.011 ± 0.024
Ti: LiTaO ₃	A _e	0.138	0.187	0.009	0.334	0.34 ± 0.04
Li _{1-2/3y} Ta _{1-2/3y} Ti _y O ₃	A _o	0.138	0.048	-0.008	0.176	0.26 ± 0.03
Zr: LiTaO ₃	A _e	(-0.009)—(-0.013)	0.122—0.115	-0.014	0.117—0.088	0.123 ± 0.015
Li _{1-2/3y} Ta _{1-2/3y} Zr _y O ₃	A _o	(-0.009)—(-0.013)	0.031—0.029	0.013	0.053—0.029	0.054 ± 0.006
Hf: LiTaO ₃	A _e	—	—	—	—	0.067 ± 0.007
	A _o	—	—	—	—	< 0.023

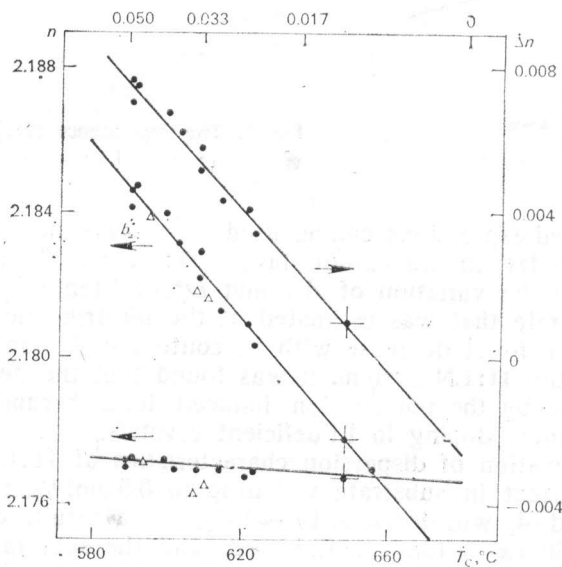


Fig. 4. The dependence of indices and birefringence of LT of T_c: a) h₀; b) h_e; c) Δn; Δ— data from [10, 11]

It should be noted, that the measurement of birefringence is in itself a fairly precise and undestructive determination of relative stoichiometry for LT once a plot such as that shown in Fig. 4 is obtained. The sensitivity of this method is ~0.1mol%Li.

The careful examination of Table.2 leads to same interesting conclusions:

— Titanium produces the most effective index increase in LT among the elements of IV sub-group.

— The decrease of A_{e,0} during the transition from Ti to Hf is induced, firstly, by the attenuation of the admixture influence on p_s, and secondly, by the reduction of the refraction variation.

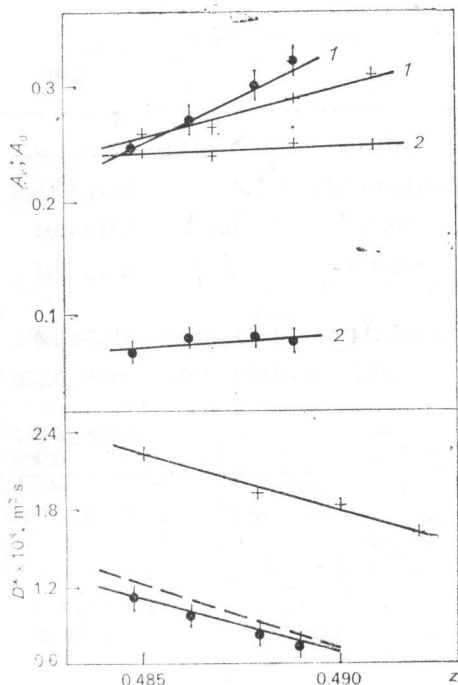


Fig. 5. The dependence of A_e , A_0 and D^* on Z :
 ● — LN; + — LT; — — — — data from [12]

The above mentioned expressions can be used to analyse the dependence of $A_{e,0}$ on the substrate stoichiometry in waveguide layers Ti:LT and Ti:LN. Figure 5 shows the results obtained on the variation of $A_{e,0}$ and D^* of titanium with molar fraction of lithium z in a substrate that was estimated at the birefringence value.

The possible reasons for A decrease with Li content depletion were analysed using the available data for the Ti:LN system. It was found that the dependence of A on z is defined on the whole by the polarization induced term because of the slowed-up decrease of T_c by titanium doping in Li deficient crystals.

The numerical estimation of dispersion characteristics of Ti:LT waveguides shows, that, if the lithium content in substrate will drop to 0.5 mol%, the Ti diffusivity will increase to $\sim 21\%$ and A_e will decrease to $\sim 19\%$. As a results of this, the difference between the effective index of fundamental mode and the substrate index will change to $\sim 30\%$, with a dramatic impact on the device performance through the change of mode size in the waveguides.

Thus, in this work we have obtained the data set that are needed for the fabrication of Ti:LT waveguides with predetermined properties. It was demonstrated, that the stoichiometry of crystal substantially affects the reproducibility of the waveguide characteristics.

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