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Analysis of nonlinear susceptibility in ion-exchanged glass

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Abstract. We demonstrate that electric-field-assisted ion-exchange in glass allows one to obtain a structure with close to a step-like concentration and conductivity profiles. Application of DC voltage to the ion-exchanged glass results in charge accumulation. The electric field induces symmetry breaking and provides quadratic optical susceptibility in the glass. Nonlinear optical properties of modified glass are studied using the Maker fringe technique. Particularly, we study the influence of the nonlinear susceptibility tensor components ratio $\chi_{zxx} : \chi_{zzz}$ on resulting characteristics. Comparing modeling and experiments, we deduced the ratio 1:3 which corresponds to Kleinman symmetry of isotropic materials.

Keywords: glass, ion-exchange, second harmonic generation, EFISH phenomenon, phase-matching

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Анализ нелинейной восприимчивости в ионообменном стекле

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Аннотация. Продемонстрировано, что ионный обмен при помощи электрического поля в стекле позволяет получить структуру с профилями концентрации и проводимости, близкими к ступенчатым. Постоянное напряжение, приложенное к ионообменному стеклу, приводит к накоплению заряда, электрическое поле которого вызывает нарушение симметрии и разрешает квадратичную оптическую нелинейность в стекле. Нелинейно-оптические свойства модифицированного стекла исследованы с использованием метода полос Мейкера. В частности, исследовано влияние соотношения компонент тензора квадратичной оптической восприимчивости $\chi_{zxx} : \chi_{zzz}$ на нелинейно-оптические характеристики. Сравнение экспериментов и моделирования позволило заключить, что это соотношение составляет 1:3, что хорошо соответствует условиям симметрии Клейнмана для изотропных сред.

Ключевые слова: стекло, ионный обмен, генерация второй гармоники, явление EFISH, фазовый синхронизм

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Introduction

Isotropic nature of glass forbids second order optical nonlinearity, limiting glass functionality in nonlinear optics. Recently, a method was demonstrated [1] that allows for breaking the material's isotropy by applying DC voltage to ion-exchanged (IE) glass at room temperature. During IE, the subsurface layer of glass is enriched with potassium ions, which provides conductivity gradient in the structure. Under applied voltage, nonequilibrium charge can accumulate in the region with conductivity gradient (Maxwell-Wagner effect). Electric field of this charge combined with the third-order nonlinearity induces effective quadratic susceptibility $\hat{\chi}^{(2)}$ due EFISH (Electric-Field Induced Second-Harmonic) phenomenon [2].

Nonlinear optical properties of the material are determined by the $\hat{\chi}^{(2)}$ tensor components, particularly, by their ratio. A powerful tool for evaluating this ratio is the analysis of Maker fringes [3] – dependence of the SH intensity ($I_{2\omega}$) on the angle of incidence of the fundamental wave (θ).

In this study, we analyze the ratio of the tensor components of the second-order nonlinear optical susceptibility of charged field-assisted ion-exchanged (FIE) glass. Advantage of the FIE is that it allows us to obtain a step-like concentration profile, where one can consider known analytical solutions applicable to crystals and thin films. Thus, having determined all parameters except the ratio $\chi_{zxx} : \chi_{zzz}$, we consider a series of Maker fringes with different thicknesses of the nonlinear layer.

Materials and Methods

In our experiments we subjected 1-mm-thick soda-lime glass slide with concentration $C_0 = 6.9 \cdot 10^{27}$ m^{-3} of mobile alkali ions to FIE. On the anodic side of the specimen, we placed a stainless-steel tube with an inner diameter of 16 mm restraining potassium nitrate melt. We use Teflon spacer between the tube and the specimen to prevent any leakage of the melt. Then we placed it in a graphite crucible with glass spacers where cathodic side of the glass contacts with $NaNO_3$ melt. The spacers create a gap filled with the melt, which allows free displacement of sodium ions from the glass by potassium ones. Then, we heated the cell up to 365 °C, applied a DC voltage of $U = 10$ V and registered DC current, which allows to calculate passed charge q_0. After, we cooled down the cell under applied voltage for about 15 min. The charge value allows us to estimate interface depth as $L = q_0/(SeC_0)$ considering full replacement of sodium by potassium, where S is the tube opening and e – electron charge.

For optical measurements we fixed the specimen in motorized rotating platform and irradiated it with a p-polarized light beam of a femtosecond laser (wavelength $\lambda = 1.03$ μm). The details of optical setup can be found elsewhere [4].

Results and Discussion

We rewrite analytical expression of Maker fringe [3] as:

$$I_{2\omega} \sim \tilde{\chi}_{\text{eff}}^{2} t'^{4}_{\omega} T''_{2\omega} \sin^2\left(\frac{1}{2}\Delta k L\right); \tag{1}$$

where $\tilde{\chi}_{\text{eff}}$ – effective nonlinear coefficient (or their combination) corresponding to the geometry of the experiment normalized by χ_{zzz}:

$$\tilde{\chi}_{\mathrm{eff}} = 2\frac{\chi_{zxx}}{\chi_{zzz}}\cos\theta'_{\omega}\sin\theta'_{\omega}\cos\theta'_{2\omega} + \frac{\chi_{zxx}}{\chi_{zzz}}\cos^2\theta'_{\omega}\sin\theta'_{2\omega} + \sin^2\theta'_{\omega}\sin\theta'_{2\omega}; \qquad (2)$$

$t'_{\text{ù}}$ – Fresnel transmission of fundamental *p*-polarized wave:

$$t'_{\omega} = \frac{2\cos\theta}{n_{\omega}\cos\theta + \cos\theta'_{\omega}}; \qquad (3)$$

$T''_{2\omega}$ – SH transmission determined as [3]:

$$T''_{2\omega} = 2n_{2\omega}\cos\theta'_{2\omega}\frac{\left(n_{\omega}\cos\theta + \cos\theta'_{\omega}\right)\left(n_{2\omega}\cos\theta'_{\omega} + n_{\omega}\cos\theta'_{2\omega}\right)}{\left(n_{2\omega}\cos\theta + \cos\theta'_{2\omega}\right)^3}; \qquad (4)$$

and Δk – wavevector mismatch:

$$\Delta k = \frac{4\pi}{\lambda}\left(n_{2\omega}\cos\theta'_{2\omega} - n_{\omega}\cos\theta'_{\omega}\right), \qquad (5)$$

θ'_{ω}, $\theta'_{2\omega}$ – angle of the propagation of the fundamental and SH waves in glass, defined by Snell's law, refractive index of these waves. For the simulations we use n_{ω} = 1.5134 and $n_{2\omega}$ = 1.5288.

Changing $\chi_{zxx} : \chi_{zzz}$ ratio from 1:2 to 1:4 only slightly modifies the shape of the Maker fringe, as illustrated in Fig. 1, *a*.

Moreover, the influence of $\chi_{zxx} : \chi_{zzz}$ on angular maxima is most noticeable for small thicknesses of the nonlinear layer, becoming negligible when the thickness approaches the coherence length, as shown in Fig. 1, *b*. Therefore, direct identification of the ratio is challenging, especially

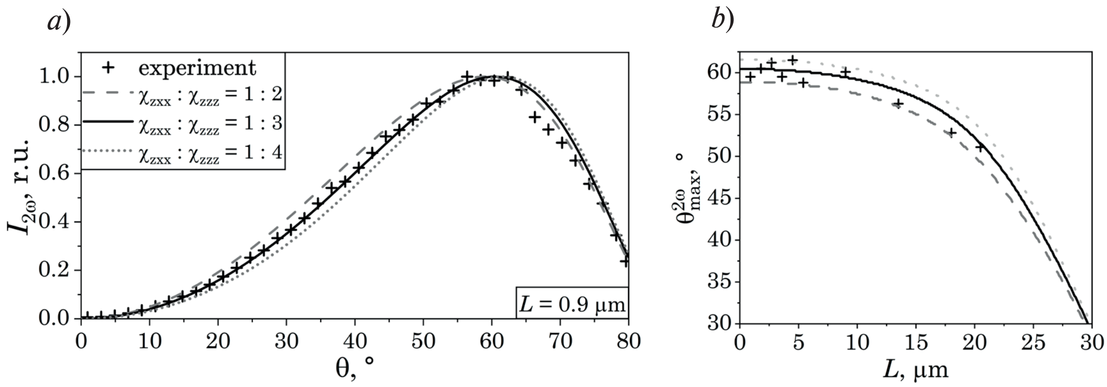

Fig. 1. Simulation of Maker fringe for L = 0.9 μm (*a*) and angle of maximum SH dependence on thickness (*b*) for different $\chi_{zxx} : \chi_{zzz}$ vs experimental data (crosses)

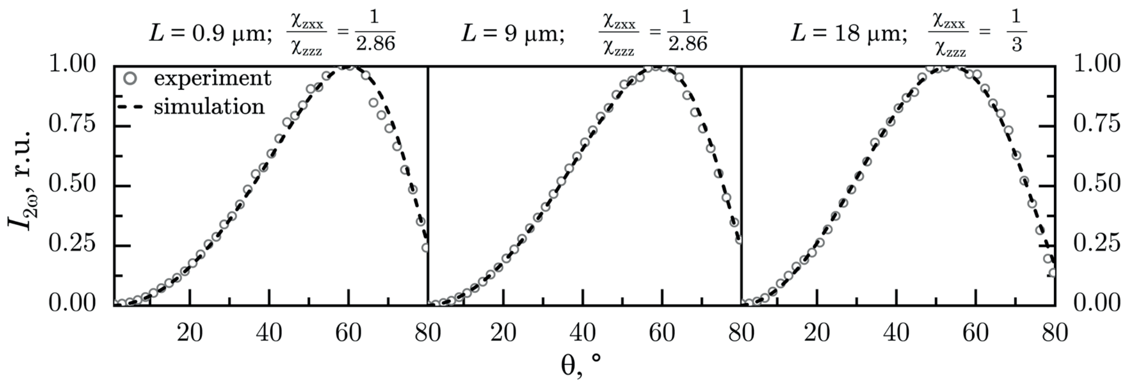

Fig. 2. Experimental data (dots) and simulation with fitted ratio of Maker fringes for L = 0.9 μm, 9 μm and 18 μm

for thicknesses close to the coherence length. For more accurate identification of the ratio we consider the ratio $\chi_{zxx} : \chi_{zzz}$ as a fitting parameter and simulate Maker fringes for the specimens different depths of FIE-modified layers. Several results of the fitting are presented in Figure 2.

The average $\chi_{zxx} : \chi_{zzz}$ for 10 specimens with L from 0.9 μm to 20.5 μm was found to be 1:3.1 which is very close to the 1:3 ratio predicted by Kleinman's symmetry condition for isotropic materials [5].

Conclusion

We analyzed the shape of angular dependences of the second harmonic in FIE glass with varying thicknesses of the ion-exchanged layer and ratio $\chi_{zxx} : \chi_{zzz}$. We determined this ratio by fitting, and obtain a value that approximately corresponds to the Kleinman symmetry condition in isotropic media. Thus, we have shown that in polarized ion-exchanged glasses the ratio of nonlinear optical susceptibility components $\chi_{zxx} : \chi_{zzz}$ is confirmed to be 1:3.

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