Deep-ultraviolet interband photorefraction in lithium tantalate

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Interband photorefraction in near-stoichiometric lithium tantalate is demonstrated and investigated at the deep-ultraviolet (UV) wavelength \( \lambda_{UV} = 257 \) nm. Formation of two distinct grating components is directly observed in depth-resolved measurements. The diffraction efficiency of a Bragg grating is measured as a function of the UV light intensity, the grating spacing, and the depth of the readout beam beneath the crystal surface. Typical time constants for the interband effects are of the order of a few tens of milliseconds for UV light intensities of approximately 100 mW/cm\(^2\), 3 orders of magnitude faster than the time constants reported previously for lithium tantalate. © 2004 Optical Society of America

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1. INTRODUCTION

Lithium tantalate \([\text{LiTaO}_3(\text{LT})]\) is widely used for acousto-optics, electro-optics, integrated optics, nonlinear optics, and holographical data storage. It has similar electro-optic coefficients as lithium niobate but is transparent over a larger wavelength range \((0.28–5.5 \text{ m})\). It was shown recently that near-stoichiometric LT (SLT) exhibits an increased photorefractive gain compared with congruent-melt LT (CLT)\(^1,2\) and is therefore of particular interest. The control of the stoichiometry of LT also leads to an increased photosensitivity of this material and to a shift of the absorption edge toward the UV. The latter is at approximately 260 nm for nearly stoichiometric crystals and at approximately 275 nm for congruent ones.\(^3\)

First experiments on the conventional photorefractive effect in SLT with the near-UV wavelength \( \lambda = 364 \text{ nm} \) have been performed previously.\(^2\) For SLT, the two-wave-mixing gain coefficient was found to be increased by a factor of 2 as compared with CLT, and photorefractive buildup and decay time constants are five times faster, for instance, 40 s for SLT compared with 200 s for CLT, both values measured for an UV intensity of 100 mW/cm\(^2\). SLT also exhibits a pronounced reduction of the saturated space-charge field, which results from a larger increase in the photoconductivity relative to the photogalvanic effect.\(^4\)

In this paper we extend the investigations of photorefractive effect in SLT to the deep-UV wavelength \( \lambda_{UV} = 257 \text{ nm} \), and, as will be shown, we observe for the first time, to our knowledge, interband photorefractive effects in this material. The photoconductivity of SLT at this wavelength was determined, and Bragg diffraction was investigated. The formation of two distinct grating components, which are most likely associated with free carriers on the one hand and deep traps on the other hand, was observed in depth-resolved measurements. We show further that the photorefractive response time is significantly reduced as compared with that of congruent samples, as well as that of SLT samples investigated at \( \lambda = 364 \text{ nm} \). The robustness of interband gratings with respect to illumination of high-power visible light is demonstrated.

2. INTERBAND PHOTOREFRACTIVE EFFECT

In analogy to the conventional photorefractive effect,\(^5\) in interband photorefraction\(^6,7\) an inhomogeneous illumination produces a correspondingly inhomogeneous space-charge distribution, which, on its side, leads to an inhomogeneous change of the refractive index of the material via the linear electro-optic effect. In contrast to the conventional case, however, the photoexcitation of charges does not occur between impurity levels and one of the conducting bands but directly across the bandgap. Such band-to-band phototransitions lead to much faster photorefractive response times \( \tau \), as was shown previously (for instance, \( \tau = 10 \mu \text{s} \) at an intensity of 1 W/cm\(^2\)) in potassium niobate (K\text{NiO}_3).\(^7\) Holographic gratings recorded in such a way are also very robust with respect to intense illumination at subbandgap wavelengths. However, ow-
ing to the large absorption of light near the bandgap, interband gratings exist only in a thin layer (typically, approximately 100–200 μm) beneath the illuminated crystal surface.

Interband photorefraction has been successfully implemented for a variety of applications, such as optical incoherent-to-coherent converters,8 optical joint Fourier-transform correlators,9 fast dynamical light-induced waveguides,10 and tunable optical filters for wavelength division multiplexing.11

3. EXPERIMENT

LT crystals grown by the conventional Czochralski method are intrinsically nonstoichiometric owing to a shift of the melt composition toward the Ta excess composition side from the initial stoichiometric composition. Conventional CLT crystals should therefore contain a considerably large amount of Ta antisite defects and cation vacancies, in analogy to the nonstoichiometric defect model of LiNbO3.13 Consequently, to grow SLT crystals, one must keep the melt composition Li rich. This is achieved by adding stoichiometric powder to the melt at a rate equal to the rate of crystal growth. This double-crucible Czochralski method was originally developed for the production of stoichiometric LiNbO3,14 and has recently been applied to LT. Crystals were grown from a Li-rich melt (58 mol.% LiO2) at a seeding temperature of 1525 °C. The growing technique and growing conditions are described in detail elsewhere.2,15

The degree of stoichiometry of the grown crystals can be estimated by measuring the Curie temperature $T_c$ for the high-temperature paraelectric-to-ferroelectric phase transition as this temperature depends strongly on stoichiometry. The Curie temperature of congruent crystals is $T_c = 601 \pm 1 ^\circ C$, and for sintered powders of exact stoichiometric composition $T_c = 692 \pm 1 ^\circ C$. By assuming a linear relationship between $T_c$ and the crystal composition, one can estimate the degree of stoichiometry that is given by the Li/(Ta + Li) atomic ratio and is ≈48.5% for congruent-melt crystals and 50% for perfect stoichiometric crystals.15 Two SLT crystals and one CLT crystal were investigated in the course of this paper that exhibit slightly different degrees of stoichiometry. Their properties are listed in Table 1.

Optical absorption spectra were measured by use of a Perkin-Elmer spectrophotometer. The effects of multiple Fresnel reflections were taken into account by use of a temperature-independent Sellmeier equation.16,17 Figure 1 depicts the absorption constant of the investigated SLT crystals as well as the spectrum of the CLT crystal in the UV wavelength range. The absorption-edge wavelength of the SLT crystals extends approximately 10 nm further toward the UV as compared with the congruent crystal. The degree of stoichiometry not only determines the UV absorption edge but also influences the absorption characteristics in the UV. Figure 1 shows that the SLT-2 crystal with its higher Curie temperature exhibits a lower absorption in the UV compared with SLT-1, and both SLT crystals absorb less in the UV than CLT. Further, the degree of stoichiometry influences the anisotropic behavior of the absorption constant with respect to light polarization. Whereas CLT is dichroic for wavelengths from approximately 285 to 370 nm, the absorption constant of SLT does not depend significantly on the light polarization in that wavelength range.

For all measurements the UV light was provided by external-cavity frequency doubling (WaveTrain, Spectra-Physics) of the output of an Ar-ion laser (Innova 200, Coherent, Inc.) operating at $\lambda = 514$ nm. Continuous-wave laser radiation at $\lambda_{UV} = 257$ nm was produced in this way with a maximum output power of approximately 100 mW.

Photorefractive Bragg diffraction experiments were performed in either a longitudinal or transverse geometry as illustrated in Fig. 2. In the longitudinal geometry, the recording beams and the read out beam enter the crystal through the same surface in a plane parallel to the $bc$ plane as shown in Fig. 2(a). As mentioned earlier, the grating that is created by the two UV beams extends only

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**Table 1. Crystals Investigated in this Paper**

<table>
<thead>
<tr>
<th>Crystal</th>
<th>Dimensions $a \times b \times c$ (nm$^3$)</th>
<th>Curie Temperature $T_c$ (°C)</th>
<th>Li/(Ta + Li) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CLT</td>
<td>$7.3 \times 1.0 \times 10.2$</td>
<td>605 ± 1</td>
<td>48.6</td>
</tr>
<tr>
<td>SLT-1</td>
<td>$10.1 \times 1.85 \times 10.0$</td>
<td>675 ± 1</td>
<td>49.7</td>
</tr>
<tr>
<td>SLT-2</td>
<td>$2.0 \times 7.9 \times 8.2$</td>
<td>684 ± 1</td>
<td>49.9</td>
</tr>
</tbody>
</table>

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**Fig. 1.** Wavelength dependence of the absorption constant $\alpha$ of the near-stoichiometric LT crystals and of the congruent sample. $\alpha_a$ and $\alpha_c$ indicate the absorption constants for light polarized parallel to the $a$ axis and $c$ axis of the crystal, respectively.

**Fig. 2.** Experimental setup (schematic) and crystal orientation for Bragg diffraction measurements in (a) longitudinal geometry and (b) transverse geometry.
approximately 150 \mu m below the crystal surface for the UV intensities investigated. Thus only a short interaction length of the readout beam with the grating is achieved.

In the transverse geometry, as depicted in Fig. 2(b), the readout beam is propagating parallel to the input surface of the recording beams. This leads to a longer interaction length, which is limited only by the size of the crystal.

4. RESULTS AND DISCUSSION
A. Absorption Constant at a Wavelength \( \lambda_{UV} = 257 \text{ nm} \)

First, we determined the absorption constants of SLT at \( \lambda_{UV} = 257 \text{ nm} \). Owing to the very large absorption of the UV radiation and the thickness of our SLT samples, the absorption constant at \( \lambda_{UV} = 257 \text{ nm} \) was measured by an indirect method. The measuring technique exploits the fact that UV illumination leads to a small reversible absorption change at visible wavelengths, which is of the order of \( \Delta \alpha \approx 15 \text{ cm}^{-1} \). A visible probe beam (wavelength \( \lambda_{\text{HeNe}} = 632.8 \text{ nm} \), intensity \( I_{\text{HeNe}} = 80 \text{ W/cm}^2 \)) propagating parallel to the top crystal surface was focused inside the SLT-2 crystal to a waist of 30 \mu m. The beam was set to propagate at some arbitrary depth \( d_1 \), as depicted in Fig. 3. The transmitted intensity of the probe beam after the crystal was monitored by a photodiode. The top surface was then homogeneously illuminated with UV light of intensity \( I_1 \), which leads to an induced transmission change, i.e., a decrease of the transmission intensity in the red. The magnitude of this decrease was registered. After this, the probe beam was moved to a different depth \( d_2 \), and the UV intensity was adjusted accordingly to the value \( I_2 \) to obtain the same change in transmission as before on illumination. By assuming that the relationship between the induced absorption change and UV intensity is a monotonic function, we can then calculate the UV absorption constant, which is assumed to be independent of intensity, with

\[
\alpha_{UV} = \frac{1}{d_2 - d_1} \ln \left( \frac{I_2}{I_1} \right). \quad (1)
\]

Note that this method does not require exact knowledge of the relationship between UV intensity and visible absorption change. Using this technique, by averaging over several measurements, we determined the absorption constant of SLT at 257 nm as \( \alpha_{UV} = 450 \pm 70 \text{ cm}^{-1} \) for light polarized parallel to the \( c \) axis and \( \alpha_{UV} = 170 \pm 30 \text{ cm}^{-1} \) for light polarized normal to the \( c \) axis.

B. Photoconductivity

To investigate the generation of free charge carriers necessary for the photorefractive effect, we measured the photocurrent as a function of the illuminating UV intensity. From this measurement, the photoconductivity was determined.

Silver electrodes were painted on the \( c \) surfaces of the SLT-2 crystal, and the electric field was directed parallel to the positive \( c \) axis. The sample was then homogeneously illuminated with UV light (\( \lambda_{UV} = 257 \text{ nm} \)) polarized parallel to the \( c \) axis of the crystal while the electric field was increased from zero to 2 kV/cm. We detected the photocurrent response by measuring the voltage over reference resistors by means of a multimeter (Keithley 195) having a large input impedance (1 G\Omega). This measurement was repeated for different UV light intensities. Tests in the absence of any external electric field showed that the pyroelectric current was negligible compared with the photocurrent. The photogalvanic current is approximately \( I_{PG} = 2.4 \times 10^{-11} \text{ A} \) at an UV light intensity \( I_{UV} = 5 \text{ mW/cm}^2 \).

Figure 4 shows the photoconductivity as a function of the UV light illumination obtained from these measurements. Owing to the high absorption of the UV light and the thickness of the sample (1.97 mm), the photoconductivity has been calculated after numerical differentiation according to

\[
\sigma_{ph} = -\frac{a I_{UV}}{b E} \frac{\Delta I_{el}}{\Delta I_{UV}}, \quad (2)
\]

where \( I_{el} \) is the current induced by illumination with light of intensity \( I_{UV} \), \( b \) is the crystal thickness in the \( y \) direction, and \( E \) is the applied electric field along the direction of spontaneous polarization of the crystal (\( z \) axis).

The photoconductivity depicted in double-logarithmic scale in Fig. 4 follows a linear dependence for low intensities.
sities, smaller than approximately 10 mW/cm². In the higher-intensity regime, an increase of photoconductivity approximately proportional to the square root of light intensity is observed as expected in the case of interband photorefraction. This suggests that for high UV intensities deep traps are not playing a major role, whereas their influence becomes evident at low intensities where the photocurrent grows linearly. Although a square-root dependence may also occur for quadratic recombination between bands and traps, this contribution can be neglected if the concentration of the traps is small as in the case of nominally pure crystals. The transition from the linear to the nearly square-root dependence takes place at an intensity \( I_s \approx 12.5 \text{ mW/cm}^2 \). We may conclude that at \( \lambda_{UV} = 257\text{-nm} \) interband photorefraction is the dominant mechanism for light intensities well above \( I_s \). As will be shown later, the value of \( I_s \) is consistent with the transition intensity obtained from measurements of the time dynamics.

The dark conductivity at room temperature was measured after preheating of the sample for 1 h at 180 °C. The value was found to be \( \sigma_{\text{dark}} = 1.5 \times 10^{-15} \Omega^{-1} \text{ cm}^{-1} \). This value was used to calculate the dielectric relaxation time

\[
\tau_{\text{dielect}} = \frac{\varepsilon_0 \varepsilon_r}{\sigma_{\text{dark}}} = 45 \text{ min},
\]

where \( \varepsilon_r \approx 46 \) is the permittivity of SLT and \( \varepsilon_0 \) is the vacuum permittivity. The dark conductivity \( \sigma_{\text{dark}} \) was found to be independent of whether the sample was illuminated by UV light or not after the heating step but prior to the measurements. Using the results of the photocurrent measurements, we may introduce a figure of merit for the photoconductivity and photorefractive response, \( \text{FM} = \phi \mu \tau_R \), where \( \phi \) is the quantum efficiency, \( \mu \) is the mobility of the charge carriers (e.g., electrons), and \( \tau_R \) is the average carrier lifetime. We assume \( \phi \approx 1 \) for interband photoexcitation transitions. The average carrier concentration is given by \( n = \phi g I_{\text{UV}} \mu \), where \( g = \alpha_{UV} / h \nu \) is a photogeneration rate and \( h \nu \) is the photon energy. By using the unipolar conductivity \( \sigma_{\text{ph}} = e n \mu \), where \( e \) is the unit charge, and neglecting the dark current, one gets

\[
\text{FM} = \phi \mu \tau_R = \frac{\sigma_{\text{ph}} h \nu}{e \alpha_{UV} I_{\text{UV}}},
\]

Note that FM is constant in the region where the photoconductivity increases linearly with intensity but FM decreases at larger intensities \( (I_{\text{UV}} > I_s) \) because \( \tau_R \) is proportional to \( (I_{\text{UV}})^{-1/2} \) in that region. In the linear region, with \( \alpha_{UV} = 450 \text{ cm}^{-1}, (h \nu) = 4.82 \text{ eV}, \) and \( \sigma_{\text{ph}} = 2.5 \times 10^{-10} \Omega^{-1} \text{ cm}^{-1} \) at an intensity \( I_{\text{UV}} = 10 \text{ mW/cm}^2 \), we obtain from Eq. (4) \( \text{FM} \approx 270 \times 10^{-16} \text{ m}^2/\text{V} \). This can be compared with \( \text{FM} = 2.6 \times 10^{-16} \text{ m}^2/\text{V} \) in congruent lithium tantalate, \( \text{FM} = 0.03 - 0.6 \times 10^{-16} \text{ m}^2/\text{V} \) in lithium niobate \( (\text{LiNbO}_3) \), and \( \text{FM} = 10 - 10^{5} \times 10^{-16} \text{ m}^2/\text{V} \) in potassium niobate \( (\text{KNbO}_3) \).

C. Photorefractive Measurements

The interband photorefractive effect in SLT is investigated by Bragg diffraction experiments in a conventional nondegenerate four-wave-mixing configuration in either the longitudinal or transverse configuration as shown in Fig. 2. The weak probe beam \( (\lambda_{\text{HeNe}} = 632.8 \text{ nm}) \) has a photon energy \( (h \nu = 2.0 \text{ eV}) \) smaller than necessary for band-to-band transitions and does not influence the interband grating.

The diffraction efficiency of the Bragg gratings was determined as a function of the writing beam intensity, the depth of the readout beam beneath the illuminated surface, and the grating constant. Further, the time dynamics of the holographic gratings were studied, as well as the influence of an additional, simultaneous intensive illumination by visible light of wavelength \( \lambda = 514 \text{ nm} \).

1. Ultraviolet Intensity Dependence

The dependence of the Bragg diffraction efficiency on the intensity of the UV writing beams was studied in the longitudinal geometry by use of crystal SLT-2 as shown in Fig. 2(a). The holographic grating is recorded symmetrically by two interfering \( p \)-polarized beams at \( \lambda_{UV} \) with a mutual angle of \( 2 \theta_{UV} = 15.6^\circ \), leading to a grating spacing of \( \Lambda = 0.95 \mu\text{m} \). The intensity of the recording beams was changed by means of UV neutral-density filters. The readout beam was focused by a lens with focal length \( f = 500 \text{ mm} \) onto the crystal and had an intensity of \( I_{\text{HeNe}} = 20.5 \text{ W/cm}^2 \) at the focal plane. The power of the diffracted beam was measured by a photodiode. As can be seen in Fig. 5, for UV intensities above \( \approx 0.1 \text{ mW/cm}^2 \), the square root of the diffraction efficiency (defined as the ratio between diffracted and incident light intensities) is approximately proportional to

\[
\eta \approx \frac{\Delta n}{\alpha} \ln \left( \frac{I}{I_{\text{ref}}} \right),
\]

where \( \Delta n \) is the average amplitude of the refractive-index change, \( \alpha \) is the absorption constant for the UV light, \( I \) is the incident UV intensity, and \( I_{\text{ref}} \) is a reference intensity needed for normalization. Such a behavior is predicted by assuming that an increase in the UV recording intensity leads to an increase of the grating’s effective thickness. When the UV light intensity is decreased beyond \( \approx 0.1 \text{ mW/cm}^2 \), the intensity of the Bragg diffraction increases.

Fig. 5. Square root of the Bragg diffraction efficiency in the longitudinal geometry as a function of the intensity of the UV writing beams for a SLT crystal with thickness \( d = 2 \text{ mm} \). The dashed line represents the best fit according to relation (5). The scale on the right indicates the corresponding diffraction efficiency.
again and does not follow relation (5). This surprising behavior can be understood by a simple model elaborated in Refs. 22 and 23 that was shown previously to apply to pure KNbO3 crystals under interband excitation. In this regime, the grating is assumed to be formed by two components that occupy different depths in the crystal and are phase shifted to each other by $\Delta \Phi$ as illustrated in Fig. 6(b).

The grating component close to the surface is induced by the space-charge modulation of free charges in the bands, having a refractive-index-change amplitude $\Delta n_5$ and thickness $d_b$. This component is referred to as the band grating. It reaches approximately from the surface to the transition depth where the intensity of the UV light equals the transition intensity $I_c$ (critical intensity dividing the linear and square-root intensity regimes of the photoconductivity). Because of the exponential decrease of the UV light inside the crystal, this transition depth is given by

$$d_b = \frac{1}{\alpha_{UV}} \ln \left( \frac{I_0}{I_c} \right).$$

(6)

The deeper grating component with refractive-index-change amplitude $\Delta n_1$ extends approximately between the point where the light intensity takes the value $I_c$ and the point where the intensity equals the dark intensity $I_d$. The latter is defined as the intensity where the induced photoconductivity equals the dark conductivity. This grating component is referred to as the trap grating as it is due to charge modulation inside deep traps. Its thickness is given approximately by

$$d_t = \frac{1}{\alpha_{UV}} \ln \left( \frac{I_d}{I_c} \right).$$

(7)

Provided that the crystal is long enough, the thickness of the trap grating is constant for UV intensities larger than $I_c$. It is just pushed deeper into the crystal for UV intensities larger than $I_c$ as the band grating expands beneath the surface.

Figure 6(b) shows a schematic view of a nondegenerate four-wave-mixing experiment involving the two grating components that was used for the measurement of the UV intensity dependence of Fig. 5. The increase of the diffracted amplitude at lower intensities gives evidence for a destructive phase shift $\Delta \Phi = \pi$ between the two gratings. Such a destructive phase shift has already been observed in interband effects in KNbO3. As discussed in more detail in Ref. 23, the phase shift can be explained by a proper unbalance between the electron and the hole modulation amplitudes at intensities below $I_c$, combined with the different mobilities proper to holes and electrons.

To summarize, the observation can be interpreted as follows. For low UV intensities, only the trap grating is existing. On increasing the UV intensity, the band grating develops and extends further into the crystal. As the band grating develops, first it decreases the total diffraction efficiency due to its opposite phase. As it grows thicker, it starts dominating the trap component until the latter can be neglected.

2. Depth-Resolved Measurement

To investigate and demonstrate the nature of the two-component grating further, we performed a depth-resolved Bragg diffraction experiment in transverse geometry. The crystal was rotated for this measurement by 90 deg with respect to Fig. 2(b) such that the UV light was incident on the $a$ facet of the crystal and the readout beam was incident on the $b$ facet. To increase the scanning resolution, we focused the readout beam along the $c$ and $b$ directions with two cylindrical lenses having focal lengths $f_c = 80$ mm and $f_b = 250$ mm. This resulted in a cylindrical beam waist of the probe beam in the center of the crystal with dimensions of approximately 34 $\mu$m in the $c$ direction and approximately 11 $\mu$m in the $a$ direction. The readout beam was adjusted to travel in the $bc$ plane. The writing beams and the readout beam were polarized nearly parallel to the $c$ axis and had intensities on entering the crystal of $I_{UV} = 140$ mW/cm$^2$ and $I_{HeNe} = 200$ mW/cm$^2$, respectively. We then measured the diffracted power while scanning the readout beam in depth.

Figure 7 shows the diffraction intensity profile as a function of the depth of the readout beam within the crystal, which correlates directly to the two distinct gratings mentioned earlier. Just beneath the illuminated crystal surface, the light is diffracted by the grating component associated with band charges; deeper in the sample, the diffraction grating is dominated by the charge modulation in the traps. With expression (6), $\alpha_{UV} = 450$ cm$^{-1}$, $I_{UV} = 140$ mW/cm$^2$, and $I_c = 12.5$ mW/cm$^2$, the thickness of the free carriers’ grating is estimated to be $d_b \approx 55$ $\mu$m, which is in good agreement with the data in Fig. 7 if one
assumes that the minimum of the diffraction efficiency at 65 µm indicates the transition region between the two gratings.

Using the result of the photoconductivity measurement (Fig. 4) and the dark photoconductivity $\sigma_{\text{dark}} = 1.5 \times 10^{-15}$ $\Omega^{-1}$ cm$^{-1}$, we can extrapolate the corresponding UV intensity to be $I_d = 0.06 \mu$W/cm$^2$. With expression (7), we can then calculate the thickness of the trap grating to be approximately $d_t \approx 270$ µm, which is in good agreement with the thickness $d_t \approx 250$ µm obtained from the depth-resolved measurement (Fig. 7).

3. Grating-Spacing Dependence

Further, the dependence of the diffraction efficiency $\eta$ on the grating spacing $\Lambda = \lambda_{\text{UV}}/(2 \sin \theta_{\text{UV}})$ was investigated in transverse geometry in the SLT-2 crystal. The total intensity of the recording beams at the crystal surface was approximately $I_{\text{UV}} = 2$ mW/cm$^2$. The readout beam was focused by a lens with focal length $f = 500$ mm onto the crystal and adjusted to travel parallel and just below the top-illuminated surface to ensure that only the grating associated with free charge carriers was probed. The intensity of the readout beam was $I_{\text{HeNe}} = 20.5$ W/cm$^2$ at the focal plane. Both the writing and the readout beams were polarized parallel to the c axis. We then varied the fringe spacing $\Lambda$ by changing the mutual angle $2 \theta_{\text{UV}}$ of the two writing beams, and the power of the diffracted beam was measured by a photodiode.

The diffraction efficiency of a photorefractive transmission grating for phase-matched Bragg diffraction in non-absorbing materials is given by

$$\eta = \sin^2 \left( \frac{\pi}{2\lambda} \left( \frac{g_p \theta_s}{\cos \theta_p \cos \theta_s} \right)^{1/2} (n_p n_s) \right)^{2/3} \frac{E_{\text{eff}}}{E_{\text{D}}} \frac{d}{D},$$

where $\lambda$ is the vacuum wavelength of the diffracted beam, $g_p$ and $g_s$ are the projection cosines of the wave-vector directions $\mathbf{k}_p = g_p/|\mathbf{k}_p|$ and $\mathbf{k}_s = g_s/|\mathbf{k}_s|$ of the pump and signal beams to the energy propagation directions $\mathbf{u}_p$ and $\mathbf{u}_s$ (walk-off angles), respectively, $\theta_p$ and $\theta_s$ are the angles between the Poynting vector of the pump and the signal beams and the sample normal, respectively, $n_p$ and $n_s$ are the refractive indices as seen by the corresponding wave, $r_{\text{eff}}$ is the effective electro-optic coefficient, $E_{\text{eff}}$ is the steady-state space-charge electric field, and $d$ is the thickness of the transmission hologram. All angles are internal to the crystal.

In our experiment, $\lambda = 632.8$ nm; $g_p$, $\theta_s \approx 1$; $\theta_p = \theta_s = \theta_{\text{Bragg}}$, where $\theta_{\text{Bragg}} = \sin^{-1} \left( \frac{\lambda_{\text{UV}}}{2 n L} \right)$; $n_p \approx n_s = n = 2.18$; and $d = 7.9$ mm is the thickness of the crystal. In a free-carrier model, in the case of an impurity-free crystal, and assuming that the mobility of one carrier species, either electrons or holes, is small enough so that the corresponding recombination field $E_{\text{R}}$ dominates, the space-charge field can be written as

$$E_{\text{sc}} = \frac{E_{\text{D}} E_{\text{R}}}{(E_{\text{D}} + 2 E_{\text{R}})(E_{\text{D}} + E_{\text{q}})}.$$

where $E_{\text{D}} = K k_B T e / \Lambda$ is the diffusion field (with $K = 2 \pi / \Lambda$, the magnitude of the grating vector; $k_B$, the Boltzmann constant; $T$, the absolute temperature, and $e$, the absolute value of the elementary charge), $E_{\text{R}} \propto \Lambda$ is the largest of the recombination fields, and $E_{\text{q}} \propto \Lambda$ is the limiting field that gives the maximum electric field that can be created by the free charge carriers.

Within the model of conventional photorefraction, on the other hand, the steady-state space-charge electric field in the case of a diffusion-dominated charge transport is given by

$$E_{\text{sc}} = \frac{E_{\text{D}} E_{\text{q}}}{E_{\text{D}} + E_{\text{q}}},$$

where $E_{\text{q}} = (\Lambda/2\pi)e N_{\text{eff}}/\epsilon_0$ is the trap limiting field. Here $N_{\text{eff}}$ is the effective number of traps, $\epsilon_0$ is the electric constant, and $\epsilon_{\text{eff}}$ is the effective dielectric constant. As can be seen in Fig. 8, taking into account Fresnel losses, a peak diffraction efficiency of 50% has been achieved for a grating spacing $\Lambda = 0.85$ µm. As the UV recording intensity was smaller than the critical intensity $I_c$, Eq. (8) was fitted to the data of Fig. 8 according to the two presented models for $E_{\text{sc}}$. In the case of the interband model described by Eq. (9), $r_{\text{eff}}$, $E_{\text{R}}$, and $E_{\text{q}}$ are used as parameters. In the case of the conventional model described by Eq. (10), the fitting parameters are $r_{\text{eff}}$ and $N_{\text{eff}}$. The dashed curve in Fig. 8 represents the theoretical curve from Eqs. (8) and (10). From the best fit we obtain $r_{\text{eff}} = 39 \pm 7$ pm/V and $N_{\text{eff}} = (3.3 \pm 0.5) \times 10^{15}$ cm$^{-3}$. The value of $r_{\text{eff}}$ is slightly larger than values known from the literature for the coefficient $r_{33}$ of LT. On the other hand, the slopes of the experimental curve are better described by the interband model according to Eqs. (8) and (9), which is indicated by the solid curve in Fig. 8. In this case, we obtain from the best fit $r_{\text{eff}} = 73 \pm 10$ pm/V, $E_{\text{R}} = 6.1 \times 10^{11}$ V/m$^2$, and $E_{\text{q}} = 3.3 \times 10^{11}$ V/m$^2$. Since Eq. (9) is valid, strictly speaking for an idealized perfectly impurity-free crystal, however, one has to be careful with the interpretation of these results.

4. Response-Time Measurements

The intensity dependence of the response time of the gratings was measured by Bragg diffraction in the longitudi-
nal geometry as shown in Fig. 2(a). In such an arrangement the observed response time of the diffracted wave is actually a mixture of the different response times of the various grating components. By reducing to the minimum terms, one can identify a fast component with exponential time constant \( \tau_1 \), which is attributed to the band grating and which carries approximately two thirds of the total diffracted amplitude and a slower component with exponential time constant \( \tau_2 \), which is attributed to the trap grating. We will distinguish between build-up and erasure times with the indices \( b \) and \( e \), respectively.

Figure 9 depicts different response times of the investigated crystals as a function of the UV writing intensity. The bottom curve shows the build-up time response \( \tau_{1,b} \) of the SLT-1 crystal. For an intensity of \( I_{UV} = 285 \) mW/cm\(^2\), the measured build-up time is approximately 20 ms. In the higher-intensity regime, \( \tau_{1,b} \) approaches a decrease with the square root of the intensity as predicted for interband gratings\(^7\) and indicated by the dashed line. The deviation of the data points from this dependency at lower intensities can be explained by the influence of traps, which slows down the buildup of the space-charge field and cannot be neglected in that case. The transition region is located at an UV intensity of approximately 50 mW/cm\(^2\), which is a bit larger than the critical intensity \( I_c \approx 12.5 \) mW/cm\(^2\) obtained from the photoconductivity measurement. However, since in longitudinal geometry one effectively probes the grating simultaneously at different depths, one does expect that the resulting averaging effect leads to a larger critical intensity than for the photoconductivity investigations.

The measured response times are approximately 3 orders of magnitude faster than response times reported previously for SLT. For example, at an intensity of \( I_{UV} = 100 \) mW/cm\(^2\), we measured a time constant of approximately 40 ms. The fastest time constants so far were measured for near-UV light (\( \lambda = 364 \) nm) and were approximately 40 s for the same illumination intensity.\(^2\) The response speed in SLT is also almost 1 order of magnitude faster than in CLT as can be seen in Fig. 9.

Further, the erasure times \( \tau_{1,e} \) and \( \tau_{2,e} \) of SLT-1 and SLT-2 were measured (indicated by “SLT” in Fig. 9). The obtained data were combined in each case as the difference between the two crystals was only marginal. For intensities lower than 100 mW/cm\(^2\), no unambiguous measurement of the decay time could be performed owing to strong compensation effects and oscillations of the diffracted power.

5. Influence of Nonresonant Illumination
The robustness of interband gratings with respect to intense illumination of light with subbandgap wavelength was investigated. The influence of a homogeneous green \( \text{Ar}^+ \) laser (wavelength \( \lambda_{\text{Ar}} = 514 \) nm, intensity \( I_{\text{Ar}} = 6 \) W/cm\(^2\) at the crystal surface) on the diffraction efficiency in longitudinal geometry was investigated. The holographic grating was recorded in the SLT-1 crystal with a grating spacing of \( \Lambda = 0.85 \mu m \), and the intensity of the recording beams was changed by means of UV neutral-density filters. The intensity of the red readout beam was approximately 0.5 mW/cm\(^2\), and the power of the diffracted beam was measured by a photodiode.

After switching on the nonresonant illumination, we detected an increase of the diffraction efficiency. Measurements were made for three UV writing beam intensities, namely, 45, 80, and 110 mW/cm\(^2\). The observed relative increase of the diffraction efficiency was 17%, 30%, and 90%, respectively. These observations can be explained by a partial or complete erasure of the grating component connected with deep traps. Since the green nonresonant illumination has almost no influence on the free-carrier concentration, the corresponding grating component is very robust and is not erased. The relative increase of the diffraction efficiency for increasing UV intensity is, however, not yet completely understood.

5. CONCLUSIONS
In summary, we have demonstrated and investigated for the first time, to our knowledge, interband photorefraction in near-stoichiometric LiTaO\(_3\) at the deep-UV wavelength \( \lambda_{UV} = 257 \) nm. The absorption constants at this wavelength were found to be \( \alpha_{UV} = 450 \pm 70 \) cm\(^{-1}\) for
light polarized parallel to the c axis and $\alpha_{\text{UV}} = 170 \pm 30 \text{ cm}^{-1}$ for light polarized normal to the c axis. The photoc conductivity of SLT was measured for UV intensities ranging from 2–25 mW/cm². A transition from linear to square-root dependency was observed at the critical UV intensity $I_{c} = 12.5 \text{ mW/cm}²$, which can be associated with the transition from trap-mediated to free-carrier-mediated photorefraction.

We observed the formation of two simultaneous principal grating components by depth-resolved diffraction experiments. The obtained thickness of the two gratings is in good agreement with theory. Measurements of the diffraction efficiency as a function of the UV writing beam intensity give evidence of a destructive phase shift between the two grating components. A maximum diffraction efficiency of 50% was achieved for $\lambda = 0.85 \mu\text{m}$ in transverse geometry and with an effective grating thickness of 7.9 mm. The dependence of the diffraction efficiency as a function of the grating constant can be explained in a satisfactory way by a simplified model, taking into account uniquely free carriers.

The temporal response of the interband effects was investigated, and the time constants were found to be of the order of a few tens of milliseconds, approximately 3 orders of magnitude faster than response times reported previously for stoichiometric LiTaO₃ at $\lambda = 364 \text{ nm}$ and approximately 1 order of magnitude faster than in congruent LiTaO₃ at the same wavelength of $\lambda = 257 \text{ nm}$. It was shown previously that highly Mg-doped lithium niobate crystals exhibit enhanced conventional photorefractive effects in the UV ($\lambda = 351 \text{ nm}$), e.g., higher gain, faster response, and lower noise, compared with undoped lithium niobate crystals. It will be interesting to investigate to what extent Mg doping also influences the interband photorefractive properties. For this purpose, suitable LT crystals are being prepared.

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