Interband photorefractive effects in KNbO\(_3\) induced by ultraviolet illumination

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A novel mechanism is used to record photorefractive gratings in dielectric crystals. The photon energy of near-ultraviolet recording beams is larger than the energy gap, and the absorbed photons excite electrons from the valence to the conduction band. These interband photorefractive gratings in undoped KNbO\(_3\) show a faster response than gratings recorded in the same material with visible light, i.e., below band-gap excitation. We measured a response time \(\tau = 5\ \mu s\) for an intensity \(I = 1\ \text{W/cm}^2\) and a refractive-index change of \(2 \times 10^{-6}\). In a transversal readout geometry, large diffraction efficiencies of 60\% in 7.5-mm-long crystals are demonstrated. Interband photorefractive gratings are only weakly affected by visible light.

Photorefractive materials can be used for many applications in dynamic holography, such as image amplifiers, image correlators, edge enhancers, and spatial light modulators. Photorefractive gratings are most often produced by illumination of a nonlinear crystal with laser light with photon energy smaller than the band gap, and the absorbed photons induce photoexcitation and redistribution of carriers within midgap centers.

The photorefractive response time is longer than or equal to the time needed to photoexcite a large enough number of carriers to create the space-charge field. In addition to the use of higher light intensities, the use of a crystal with a large absorption constant \(\alpha\) may also be helpful in increasing the response speed. But \(\alpha\) can be increased only in a limited range by doping. In contrast, large absorption is observed without doping at the band edge of any photorefractive material.

The creation of photorefractive gratings by photoexcitation over the band gap has been demonstrated recently in multiple-quantum-well devices. However, the thickness of such multilayer structures is limited because of the epitaxial growing process, and reasonable diffraction efficiencies are obtained only by making use of resonant nonlinear effects at the band edge. This limits the readout wavelength range to a width of 10–20 nm and the optical interaction length to a few micrometers. The use of oxide crystals with large linear electro-optic coefficients, such as KNbO\(_3\), overcomes this problem because the readout can be done at any wavelength in the visible. Even though the thickness of the photorefractive grating decreases with an increasing absorption constant, the interaction length \(L\) can be increased to the crystal size by propagating the nonabsorbed readout beams parallel to the crystal surface. In this Letter we present photorefractive experiments performed in undoped KNbO\(_3\) with UV recording beams. To our knowledge, these are the first investigations of interband photorefractive effects performed in dielectric materials.

The Ar-ion laser wavelengths \(\lambda = 363.8\) and \(\lambda = 351.1\) nm were used for recording. These are shorter than the band-edge wavelength (\(\lambda \approx 375\) nm) of KNbO\(_3\). The readout was performed by use of visible beams (\(\lambda = 632.8\) nm) in the transversal geometry shown in Fig. 1(b). In this configuration the readout beam is propagating parallel to the input surface of the recording beams. Samples with the largest faces perpendicular to the \(a\) or \(b\) axis were polished to optical quality (planarity \(= \lambda/4\)). The recording beams were always oriented in such a way that the grating wavevector was parallel to the crystal \(c\) axis, and, when necessary, electric fields were applied in the +\(c\) direction through silver-painted electrodes on the top surface.

The absorption constant \(\alpha\) of KNbO\(_3\) is anisotropic and is larger for \(b\)-polarized light than for \(a\)- or \(c\)-polarized light. The values at our laser wavelengths were determined by measurement of the transmission in thin samples and by photorefractive measurements in transversal geometry. In the second case the readout depth below the surface was varied, and the intensity of the incident UV beams was adjusted to get an unchanged diffraction efficiency or time response at each depth. The required intensities as a function of the readout depth can then be related to \(\alpha\). The values of \(\alpha\) obtained with the

![Fig. 1. (a) Longitudinal and (b) transversal recording geometry. In the transversal geometry the readout beam has an elliptical cross section with vertical half-axis of 20 \(\mu m\).](1144_141144_03S6.00/0)
At \( \lambda = 364 \) nm, the intensity \( I_0 = 0.06 \) W/cm\(^2\); readout: \( \lambda = 633 \) nm, intensity \( I = 1 \) W/cm\(^2\). The recording and readout beams are polarized parallel to the crystal’s c axis. The grating spacing is \( \Lambda = 2.8 \) \( \mu \)m, the interaction length is \( L = 7.5 \) mm, and the readout beam propagates 15 \( \mu \)m below the crystal surface.

two methods agree within the experimental errors. At \( \lambda = 364 \) nm, \( a_r = 540 \pm 50 \) cm\(^{-1}\) and \( a_t = 1000 \pm 100 \) cm\(^{-1}\), whereas at \( \lambda = 351 \) nm the values are \( a_r = 1900 \pm 150 \) cm\(^{-1}\) and \( a_t = 5500 \pm 2000 \) cm\(^{-1}\).

For the photorefractive measurements, the use of the transversal geometry of Fig. 1(b) instead of a longitudinal geometry [Fig. 1(a)] has two advantages. First, the interaction length of the readout beam with the photorefractive grating is longer, which gives larger diffraction efficiencies \( \eta \). Second, the readout depth can be varied, permitting a better characterization of the refractive-index change and its time response. Figure 2 shows a measurement of \( \eta \) as a function of the applied field \( E_0 \) performed at \( \lambda = 364 \) nm and for an interaction length \( L = 7.5 \) mm. \( \eta \) is defined as the ratio of the diffracted to the incident wave intensity inside the crystal. At the maximum the diffraction efficiency is \( \eta = 60\% \). The dashed curve in Fig. 2 is a fit to the formula

\[
\eta = R \sin^2(\pi \Delta n L / \lambda \cos \theta),
\]

\[
\Delta n(E_0) = \Delta n(E_0 = 0) + kE_0.
\]

Here \( \lambda \) is the readout wavelength and \( \cos \theta \) is the internal readout angle. The fit is obtained under the assumption that the average refractive-index change \( \Delta n \) and the space-charge field amplitude \( E_0 \) increase linearly with the field \( E_0 \). The scaling factor \( R \) is introduced to account for the fact that the maximum of \( \eta \) is not at 100\%. Because the vertical cross section of the readout beam is approximately 40 \( \mu \)m [Fig. 1(b)], we may attribute the factor \( R \) to a variation of the refractive-index change \( \Delta n \) with the depth below the crystal surface.

We note that in Fig. 2 the visible readout intensity is approximately 20 times higher than the writing intensity. Nevertheless, the grating is not erased, which is not surprising owing to the low absorption (\( \alpha \approx 0.05 \) cm\(^{-1}\)) at 633 nm. The robustness of the gratings to the readout light is even stronger and is characteristic of the interband photorefractive gratings. Preliminary experiments show that there is a component of the grating that is not erased even with the use of visible readout beams at \( \lambda = 514 \) nm that are more than 5 orders of magnitude stronger than the UV writing beams. The ratio of the absorption constants at the two wavelengths, which is approximately 3000, is smaller than the ratio of visible to UV power. Whereas the UV light predominantly induces electron transitions from the valence to the conduction band, visible light can induce transitions only between deep impurities and the bands. Some transitions of the latter kind may also be induced by UV light, but the related absorption constant is much smaller than the one for interband transitions. Thus UV and visible photons act essentially on two different charge reservoirs, the first on free carriers in the bands, the latter on charges trapped in deep impurities. Therefore the observed robustness may be due to a grating formed by free carriers whose modulated concentration is almost immune to visible excitation. The free carriers may be charges moving in the bands or may be quasi-free carriers (i.e., free carriers that are temporarily trapped in shallow levels that are in fast thermal exchange with one band).

In the transversal geometry, the fastest response is observed when the readout beam is aligned as close to the crystal surface as possible, because the local writing intensity is higher there. For experiments on the response time, the recording beams are switched on and off by an acusto-optical modulator placed before the beam splitter of the holographic setup. The switching time is approximately 0.1 \( \mu \)s. The grating is erased by switching off both recording beams derived from the first diffraction order of the modulator and simultaneously switching on a homogeneous illumination at the same wavelength (zeroth-order beam of the modulator). In Fig. 3 one sees an example of the recording–erasure cycle. A fast and a slow component, which are especially evident when one is looking at the erasure process, can be distinguished. The exponential

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\text{Fig. 2. Electric-field dependence of the diffraction efficiency measured in transversal geometry. Recording: } \lambda = 364 \text{ nm, intensity } I_0 = 0.06 \text{ W/cm}^2; \text{ readout: } \lambda = 633 \text{ nm, intensity } I = 1 \text{ W/cm}^2. \text{ The recording and readout beams are polarized parallel to the crystal's c axis. The grating spacing is } \Lambda = 2.8 \text{ \( \mu \)m, the interaction length is } L = 7.5 \text{ mm, and the readout beam propagates 15 \( \mu \)m below the crystal surface.}
\]

\[
\text{Fig. 3. Recording–erasure cycle of interband photorefractive gratings in KNbO}_3 \text{ for } \Lambda = 4.7 \text{ \( \mu \)m and } E_0 = 2.2 \text{ kV/cm. A He–Ne laser beam (} \lambda = 633 \text{ nm) is transversally reading out the 2-mm-long grating in 10-\( \mu \)m depth. For the erasure, we subtract from the measured points a single exponential function describing the slow erasure component (} \tau = 65 \text{ \( \mu \)s). The transformed data points are drawn below the original ones and give an exponential time constant } \tau = 11 \text{ \( \mu \)s for the fast component. The recording intensity is 0.35 W/cm}^2, \text{ the erasure intensity is 0.25 W/cm}^2, \text{ and the readout intensity is approximately 10 W/cm}^2.\]

the equation

traps, the average density no of free electrons fulfills bands. When we neglect all the processes involving is proportional to the free-carrier concentration, is in the same intensity range the photocurrent, which increases linearly with the free-carrier concentration. This can be explained by a quadratic carrier recombination process between the interband carrier photoexcitation and the interband carrier recombination process.

In Eq. (4), \( \varepsilon \) is the relative dielectric constant, \( \varepsilon_0 \) is the permittivity of vacuum, \( e \) is the unit charge, and \( \mu \) is the carrier mobility.

All the investigations shown above were performed in the transversal geometry shown in Fig. 1(b). We mention here that interband gratings can also be read out longitudinally [Fig. 1(a)]. In preliminary experiments we measured a diffraction efficiency \( \eta \) of \( 1.4\% (\lambda = 633 \text{ nm}) \), even for the moderate UV writing intensity of \( I_w = 12 \text{ mW/cm}^2 \). The UV beams (\( \lambda = 364 \text{ nm} \)) were polarized in the \( a-c \) plane, and an electric field \( E_0 = 5 \text{ kV/cm} \) was applied. We estimate that under the above conditions the effective grating thickness is approximately 190 \( \mu \text{m} \). We note that this value of \( \eta \) is comparable with the maximum diffraction efficiency measured in multiple quantum wells by use of ac fields and resonant readout.7

In conclusion, we have demonstrated, for the first time to our knowledge, the recording of photorefractive gratings in dielectric electro-optic crystals by using light with photon energy larger than the band gap. The grating response time is much shorter than that with the use of conventional photoexcitation mechanisms because of high absorption. We measured \( \tau = 5 \mu \text{s} \) in a transversal! recording geometry by using a UV intensity of 1 \text{W/cm}^2. In this geometry, the interaction length \( L \) of the readout beam is determined by the dimensions of the writing interference pattern or by the crystal size. A maximum diffraction efficiency of 60\% is obtained for \( L = 7.5 \text{ mm} \) with an applied field \( E_0 = 2.3 \text{ kV/cm} \). The intensity dependence of the response time and the robustness of the gratings to intense visible light suggest that free (or quasi-free) carriers play a major role in the space-charge field formation. Potential applications of interband photorefractive effects include high-speed and high-resolution spatial light modulators for parallel optical processing devices or optical correlators. Beam steering in optical waveguides also seems very promising.

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References