Photoconductivity of photorefractive Sr$_{0.61}$Ba$_{0.39}$Nb$_2$O$_6$:Ce crystals at high light intensities
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I. INTRODUCTION

Photoconductive Sr$_{0.61}$Ba$_{0.39}$Nb$_2$O$_6$ (SBN) is a promising optical material. Recently SBN crystals were investigated for various applications, e.g. for volume holographic storage, self-pumped phase conjugation, light modulation, spatial filtering, and for photoassisted switching of ferroelectric domains. Photorefractive properties and light-induced charge transport are of special interest. At pulse laser intensities new effects arise, e.g. an enhancement of the photorefractive sensitivity by acceleration of the charge carriers in pyroelectric fields. Furthermore, high intensities as an extreme case are useful to obtain detailed information on the light-induced charge transport properties. The photoconductivity plays a crucial role in the photorefractive effect. In this article we develop a special method for direct photoconductivity measurements and investigate SBN crystals doped with Ce under the influence of high intensity light pulses.

II. EXPERIMENTAL METHODS

From the time constant of holographic erasure experiments the photoconductivity can be estimated. However, this estimation always requires some assumptions, e.g. about trap densities (space charge limitations), drift lengths and secondary photorefractive centers. Thus in many cases directly measured data are more reliable. Applying a dc high voltage to a crystal and observing the resulting dc current with an electrometer, or applying an ac voltage and detecting the ac current with a lock-in amplifier are well-known techniques for the direct determination of the photoconductivity. However, in the case of illumination with short light pulses of high intensity these techniques fail because the bandwidths of electrometer and lock-in amplifier are much too small. Lesaux et al. preferred another technique. They applied a dc high voltage, too, but instead of the electrometer used an oscilloscope with a 50 Ω input resistance. The current through the oscilloscope creates a voltage which is detected. By this technique photoconductivity measurements of Sr$_{0.61}$Ba$_{0.39}$Nb$_2$O$_6$:Ce crystals were successfully carried out. But other photorefractive crystals, e.g. SBN, show a photoconductivity which is orders of magnitude smaller than that of BSO, and for this reason higher sensitivity is required. An enhancement of sensitivity by an increase of the input resistance of the oscilloscope may cause problems, because the bandwidth can be decreased too much. Thus in the case of higher insulating photorefractive crystals this method is not useful.

Here we present a new technique for direct photoconductivity measurements: A high voltage is applied to the sample and subsequent illumination yields a photocurrent through the crystal. This current charges a capacitor and finally an electrometer measures the voltage of this capacitor. From voltage, capacity and exposure time the photoconductivity is calculated.

A. Crystals

Strontium–barium niobate crystals of congruently melting composition Sr$_{0.61}$Ba$_{0.39}$Nb$_2$O$_6$ (SBN) are used in the experiments. SBN has a tungsten–bronze structure and is ferroelectric at room temperature (point group 4mm). The transition to the paraelectric phase occurs at $T_C=80 °C$. All samples are grown at the Crystal-Growth Laboratory of the Physics Department of the University of Osnabrück by the Czochralski technique. Growth temperature and pulling rate are 1480 °C and 0.7 mm/h. Doping is performed by adding different amounts of CeO$_2$ to the melt. All Ce concentrations mentioned in this paper are given in weight percent of CeO$_2$ in the melt. The crystals are cooled down to room temperature within 20 h and show a lightly red brown color which becomes darker with increasing Ce content. After cutting the samples the surfaces are polished to optical quality.
Heating of the crystals up to 110 °C and cooling them down to room temperature with an applied field of 600–800 kVm⁻¹ yields single-domain samples. In Table I dimensions of the crystals and the Ce concentrations in the melt are listed.

### B. Setup

Light pulses are generated with the help of a Q-switched frequency-doubled Nd:YAG laser. Light wavelength, pulse duration and pulse energy are about λ = 532 nm, tₚ = 20 ns and Eₚ = 150 mJ. The light pulses are directed onto the samples and pass through the crystals along the b axis (ordinary polarization). Different light intensities are obtained by attenuation of the pulses with different neutral density filters mounted on a rotatable filter holder. The beam diameter of 4 mm (1/e² width) ensures a sufficiently homogeneous illumination of the samples. A computer serves to control filter holder and pulse laser, and to acquire the data.

### C. Electronic circuit

Figure 1 shows schematically the electronic circuit used. A high voltage source charges the capacity C_measure = 47 nF through the crystal. Illumination of the crystal with light pulse yields a high photoconductivity and for a short time a large current flows through the crystal to the capacitor. Two relays are used to discharge the capacitor through R_discharge or alternatively connect an electrometer which determines the voltage U_measure of C_measure. A choke coil and a glow lamp provide an overvoltage protection of the electrometer.²² If a voltage breakdown at the crystal happens, the choke coil slows down the voltage increase and the glow lamp short circuits the electrometer at about 200 V. High voltage source, relays and electrometer work are computer controlled. A voltage U_measure much smaller than U_load ensures that the field applied to the crystal remains constant.

### D. Dark and photoconductivity

The amount of charge Q captured in C_measure results from the current through the crystal. With a completely discharged capacitor at the beginning of the measurement we can write

\[ Q = C_{\text{measure}} U_{\text{measure}} = j A t, \]

where j is the time-averaged current density through the crystal, A the electrode area of the sample and t the measuring time after completely discharging the capacitor. The field applied to the crystal is given by \( E_{\text{load}} = U_{\text{load}} / l \), where l is the electrode distance and the time averaged conductivity is given by \( \sigma = j / E_{\text{load}} \). Therefore Eq. 1 yields

\[ \sigma = \frac{U_{\text{measure}}}{U_{\text{load}}} \frac{l}{A} \frac{C_{\text{measure}}}{t}. \]

Without illumination t is the time for which a field was applied to the crystal and Eq. 2 yields the dark conductivity. For photoconductivity measurements we subtract the dark conductivity effect as a linear offset from the data U_measure(t). In the case of pulse laser illumination, t is the pulse duration tₚ and ω the photoconductivity ω_ph. If several pulses are used to accumulate charge, the voltage...
$U_{\text{measure}}(t)$ has a steplike behavior and in Eq. 2 the time $t$ is given by $t_p$ times the number of used pulses.

Figure 2 shows typical evolutions of $U_{\text{measure}}(t)$ for different applied voltages after subtraction of a linear offset. Without applied voltage, after each pulse a transient signal $U_{\text{measure}}$ appears for a few seconds. This signal is attributed to influences of the pyroelectric effect because after some time the signal $U_{\text{measure}}$ goes back to zero. The relaxation occurs with a time constant of the order of 1 min, which is typical for cooling of the crystal. Therefore it is necessary to use the signal $U_{\text{measure}}$ obtained at least 1 min after the last light pulse, in order to get reliable information about the amount of charge transmitted through the crystal.

E. Precautions

It is necessary to take into account that the electrometer may charge the capacitor $C_{\text{measure}}$ by its input. This charging leads to a signal $U_{\text{measure}} \neq 0$ even for $E_{\text{load}}=0$ and causes a linear offset in $U_{\text{measure}}(t)$, which must be always subtracted. To obtain reliable dark conductivity values measurements with different applied fields $E_{\text{load}}$ are necessary.

The fields applied to the samples may be attenuated by about 10% because of remaining intensity inhomogeneities of typically 10% over the crystal front face. The space charge distribution inside the crystal can influence the following measurements, especially if measurements with a large field precede measurements with small fields. This unwanted influence is eliminated by the following procedure: Before each measurement the crystal is exposed to white light for 5 minutes and then kept in the dark for 25 minutes.

III. EXPERIMENTAL RESULTS

A. Ohmic behavior

Figure 3 shows a typical result for the dependence of $U_{\text{measure}}$ on the applied voltages $U_{\text{load}}$. It is obvious that Ohm’s law is very well fulfilled. Furthermore, from Figure 3 we see that $U_{\text{measure}}(U_{\text{load}}=0)=0$, showing that the bulk photovoltaic effect in SBN:Ce is negligible. Fits like in Figure 3 yield the slope $U_{\text{measure}}/U_{\text{load}}$ which is required for the evaluation of the photoconductivity according to Eq. 2.

B. Dependence of photoconductivity on light intensity

The intensity dependence of photoconductivity $\sigma_{\text{ph}}$ may always be described by a relation $\sigma_{\text{ph}} \propto I^x$. Figure 4 shows an example. The light intensity values are averaged over the crystal depth taking into account reflection and absorption losses. This is of importance to enable comparison of $\sigma_{\text{ph}}$ values measured for samples of different thickness or absorption.

C. Dependence on the cerium concentration

In Figure 5 the dependence of the photoconductivity $\sigma_{\text{ph}}$ and of the nonlinearity parameter $x$ on the CeO$_2$ concentration $c_{\text{Ce}}$ in the melt is shown. The photoconductivity reaches the highest values around $c_{\text{Ce}}=0.05$ wt % CeO$_2$. For the same Ce concentration the intensity dependence becomes very sublinear (small $x$).
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Further improvements of the intensities strong nonlinearities of the photovoltaic effect are at high light intensities are of interest because at high light example, direct measurements of the bulk photovoltaic effect.

Several new applications of the method are possible. For instance, information concerning the time evolution of the conductivity can be obtained if the external electric field is applied to the crystal only for a short time. This may be realized, e.g., by connection of the crystal electrodes with two wires of different lengths. Simultaneously applying a high voltage to both ends of these wires generates an applied field which is present only for a short time; the high voltage reaches the crystal faster through the short wire than through the long one. Measurements with different delay times between pulse illumination and field application then yield $\sigma_{ph}(I)$.

### B. Integrated photoconductivity

The photoconductivity values may consist of two contributions: a photoconductivity during pulse illumination and an enhanced dark conductivity after illumination resulting from, e.g., thermal depletion of shallow levels. However, the dark conductivity effect cannot be responsible for the observed $\sigma_{ph}(I)$ nonlinearities: Earlier we found that light-induced absorption changes increase linearly with the light intensity in the intensity range used in the present investigation. This is an indication that the number of charge carriers, which may become thermally excited and which are responsible for the enhanced dark conductivity after the pulse illumination, is proportional to $I$.

### C. Influence of cerium doping

For small Ce concentrations ($c_{Ce} \approx 0.025$ wt % CeO$_2$ in the melt) Ce plays a minor role in the light-induced charge transport, but Ce becomes dominant for larger $c_{Ce}$ values. Around $c_{Ce} \approx 0.05$ wt % CeO$_2$ the photoconductivity $\sigma_{ph}$ is large and very sublinear (small $\alpha$). Both effects can be explained by a lack of traps. A similar effect was observed in investigations of SBN-Ce with continuous wave (cw) light of intensities up to 30 kW m$^{-2}$. Therefore we conclude that at cw and pulse light intensities the same photorefractive center is of importance, which has been recently identified in the cw intensity region as Ce$^{3+}$.

A two-level model may be used to explain the relationship $\sigma_{ph} \propto I^{x}$. However, if a secondary level is present, the high photoconductivity of the samples doped with $c_{Ce} = 0.025$ and 0.05 wt % CeO$_2$ should cause a pronounced population of the shallow level and should yield large light-induced absorption changes, which are not observed. Instead, the results may be explained by population of an intrinsic center by direct excitation of charge carriers from Ce$^{3+}$. A similar situation is observed in LiNbO$_3$:Fe at high light intensities. Another possible explanation is simultaneous electron and hole conductivity. Then the electrons may cause the large photoconductivity and the holes may recombine with a shallow center and cause the light-induced absorption.

For higher Ce concentrations ($c_{Ce} \geq 0.1$ wt % CeO$_2$) a nonlinearity of $\sigma_{ph}(I)$ with $x \approx 0.8$ is still present. Because the $x$ values show no further increase with increasing $c_{Ce}$, it is unlikely that in this region the sublinearity of the photoconductivity is caused by a light-induced change of the trap concentration. Probably here a two-level situation is responsible for the observed sublinear behavior of $\sigma_{ph}(I)$.

### V. SUMMARY AND CONCLUSIONS

Applying an electric field to a crystal, storing the current through the sample in a capacitor and subsequently measur-

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**FIG. 5.** Photoconductivity $\sigma_{ph}$ (intensity 20 GW m$^{-2}$) and nonlinearity parameter $\alpha$ versus $c_{Ce}$, the concentration of CeO$_2$ in the melt. The symbols represent measured results and the solid lines are guides to the eye.
ing the voltage of the capacitor is a useful technique for direct photo- and dark conductivity measurements at low and high light intensities.

Photoconductivity measurements of SBN:Ce with frequency-doubled pulses of a Q-switched Nd:YAG laser yield a sublinear intensity dependence \( \sigma_{ph} \propto I^x \) with \( x < 1 \). For Ce concentrations smaller than 0.1 wt % CeO\(_2\) in the melt the nonlinearity is probably caused by a light-induced change of the concentration of traps for the free charge carriers, while for larger Ce concentrations a two-level system seems to be responsible for the sublinearity. In SBN:Ce at high light intensities probably Ce\(^{3+}^{4+}\) is the dominant photorefractive center. Further investigations are required for a complete understanding of the processes in SBN:Ce at high light intensities, e.g. measurements of the light-induced absorption changes in heavily doped samples, determination of the two-beam coupling gain for different intensities and grating period lengths, or investigation of the time evolution of the photoconductivity.

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