Temperature dependence of the electrooptic coefficients r_{22} and m_{22} in LiNbO$_3$

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Measurements of temperature dependence of linear electrooptic coefficients defined in terms of the electric field r_{22} and the induced polarization m_{22} were made for LiNbO$_3$ within the temperature range 25°C–200°C. After annealing, the coefficient r_{22} in samples with $x_c \approx 49.3\%$ is found to be nearly temperature independent, increasing at 125°C only 0.6%. Contrary to the commonly observed temperature behaviour of intrinsic coefficients, m_{22} instead of r_{22}, changes significantly with temperature.

Keywords: electrooptic effect, LiNbO$_3$.

1. Introduction

The linear electrooptic coefficients are often noticeably temperature dependent. This feature limits applicability of some crystals in technical devices by decreasing their temperature stability. The temperature changes of electrooptic coefficients are important, for example, in high-voltage systems for which an increased interest in unconventional methods based on the electrooptic effect may be observed. This includes electric field sensors and optical voltage transformers [1–3]. Among crystals proposed for application in these devices is LiNbO$_3$, specifically its r_{22} coefficient. Thus, the linear electrooptic effect in this material is of great significance.

The aim of this work is to investigate the temperature dependence of the r_{22} coefficient in LiNbO$_3$ and its intrinsic counterpart m_{22}.

2. Measurement

In a crystal of the point symmetry 3m, the linear electrooptic coefficient r_{22} may be measured with the electric field $E = (E,0,0)$ and the light beam propagating along the optic axis or with $E = (0,E,0)$ and the light sent perpendicular to this axis. According to the results of recent calculations based on the Jones calculus we choose the latter configuration providing better experimental accuracy [4–6].

The experimental approach adopted in this work involved the dynamic polarimetric method [7,8]. The absolute value of r_{22} was evaluated from the modulation index of the emerging light intensity detected by a photodiode by means of a lock-in technique. The computer-controlled apparatus changed the temperature within the range 25°C–200°C and monitored both the dc and modulated components of the photodiode output signal. In each cycle, the subsequent temperature points were reached approximately linearly in 2 hours, then the temperature was stabilized for 1 hour before measurements at specific temperatures. Intervals between subsequent cycles lasted 2 or 3 days. At the end of each cycle, starting from 200°C the temperature was linearly decreased in 8 hours. The optical system used in these measurements was biased to the middle of its transmission characteristic by taking advantage of the temperature dependence of the natural birefringence of the measured crystal [7,8]. The investigations were made in air on the samples with dimension of 30×30×4 mm^3. The LiNbO$_3$ samples under investigation have been grown, cut and polished 8 years ago and kept during this time in sealed containers. The samples were purchased from Cobrabid-Optica Warsaw and produced at the Institute of Electronic Materials Technology, Warsaw. A He-Ne laser beam was passed along the [100] direction of the sample sandwiched between crossed polarizers. A sinusoidal electric field of frequency 417 Hz and amplitude 10 kV/m was applied to the sample. For this frequency the crystal may be assumed to be mechanically free. In our measurements we took into account the temperature dependence of the ordinary refractive index of LiNbO$_3$ from Ref. 9.

Our results showed that during the first cycle of measurements the r_{22} coefficient was noticeably dependent on temperature approaching maximum at about 125°C. We found that for next measurements the increase in r_{22} was weaker. This is illustrated in Fig. 1, where the dependencies labelled (a), (b), (c), and (d) correspond to subsequent cycles of heating. As it is shown in Fig. 1, after about 10 such cycles the r_{22} coefficient was nearly independent of temperature increasing at 125°C only 0.6% relative to room temperature. Our measurements were repeated during next months. The results obtained indicate that the effect is sta-
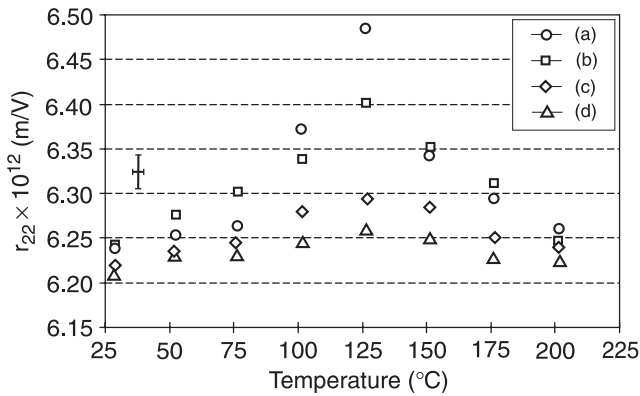
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Fig. 1. Temperature changes in the linear electrooptic coefficient r_{22} of investigated LiNbO$_3$ samples. The dependencies correspond to subsequent cycles of heating, namely: (a) the first, (b) the fifth, (c) the eighth, and (d) the tenth heating.

ble. After 9 months the changes in the r_{22} coefficient were practically the same as those labelled (d) in Fig. 1.

It is known that many properties of LiNbO$_3$, including dielectric and electrooptic ones, are strongly dependent on the composition of the crystal [10–13]. Thus the knowledge of the electrooptic coefficients and the dielectric constants allows the composition of the crystal to be determined. The composition is often described by the ratio $x_c = $ [Li]/([Li] + [Nb]). For crystals approaching the stoichiometric composition x_c equals to 50%.

The temperature changes in the low-frequency dielectric constant ε_2 measured at 417 Hz are shown in Fig. 2. We fitted this dependence by a Curie-Weiss-type formula

$$\varepsilon_2 = \varepsilon_2^\infty + C_2/(T - T_2), \qquad (1)$$

with $\varepsilon_2^\infty = 45.9$, $C_2 = 43.5 \times 10^3$ K and $T_2 = 1213$ K. We measured also the low-frequency dielectric constant ε_3. At room temperature we obtained $\varepsilon_3 = 28.7$. Comparison of our values determined for ε_2, ε_3, and r_{22} with that reported

in Refs. 10, 11, and 12 allows to estimate the composition of our samples to be about $x_c = 49.3\%$.

It was Pockels who first noticed that electrooptic and dielectric properties of crystals are closely related and that electrooptic coefficients follow the dielectric behaviour of crystal [14]. Miller has generalized the approach for second harmonic generation and linear electrooptic effect by introducing delta coefficients, which are normalized against linear susceptibilities of relevant frequencies [15]. Following Pockels we considered in our work the so-called intrinsic electrooptic coefficient m_{22} defined in terms of the induced polarization rather than the macroscopic electric field, as is the case of the r_{22}. According to its definition, r_{22} is related to m_{22} by

$$r_{22} = \varepsilon_0(\varepsilon_2 - 1)m_{22}, \qquad (2)$$

where ε_0 is the permittivity of free space. The intrinsic electrooptic coefficients are usually observed to be nearly temperature independent. However, our results presented in Fig. 3 show that for the m_{22} coefficient in LiNbO$_3$ this is not the case.

Fig. 3. Temperature dependence of the intrinsic linear electrooptic coefficient m_{22} of investigated LiNbO$_3$ samples after 10 cycles of heating.

3. Discussion

The previous measurements of the temperature dependence of the linear coefficient r_{22} in LiNbO$_3$ have been reported in Ref. 16. In temperatures below 125°C, the dependence was found to be not significant. However, in the region above 125°C an unusually strong decrease in the r_{22} coefficient has been observed. This is different from the behaviour of r_{22} in our samples.

As it is discussed in Ref. 12, anomalies of various physical properties at around 100°C have been previously reported for LiNbO$_3$. The anomalies have been supposed to be related to the nonstoichiometry of the crystals. The deviation from the stoichiometry increases the content of intrinsic defects. Thus, annealing the crystals should decrease the effect of the intrinsic defects on r_{22} what seems to explain the plots shown in Fig. 1.
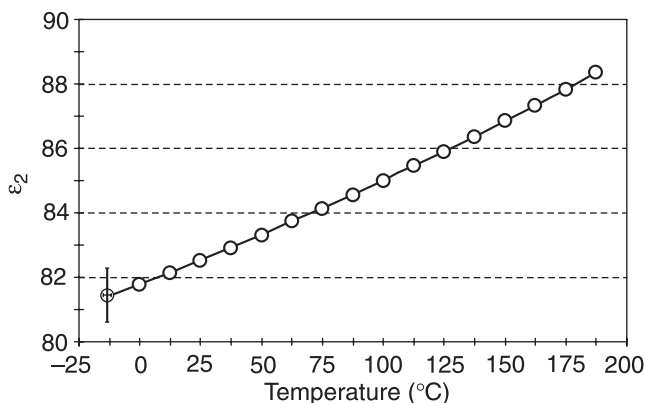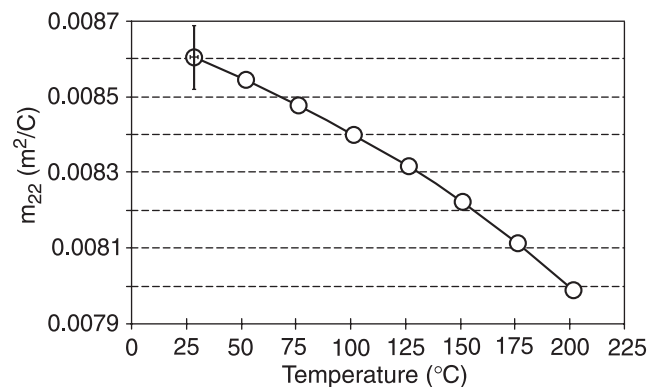

Fig. 2. Temperature dependence of the low-frequency dielectric constant ε_2 of investigated LiNbO$_3$ samples after 10 cycles of heating.

Attempts to relate various components of the linear electrooptic tensor in LiNbO$_3$ to the quadratic electrooptic response of the oxygen octahedra of the equivalent centrosymmetric state have been previously presented [17]. The linear electrooptic effect has been considered as a morhpic effect induced by the spontaneous polarization. In this approach, the non-zero value of r_{22} results from the distortions of the octahedra. It has been found that the changes in the composition of LiNbO$_3$ are related with these distortions [11]. The weak temperature dependence of r_{22} suggests that the distortions of the oxygen octahedra are more sensitive for the crystal composition than its temperature.

4. Conclusions

After annealing, in our investigated LiNbO$_3$ crystals with $x_c \approx 49.3\%$, the linear electrooptic coefficient r_{22} is within temperature range between 25°C and 200°C nearly temperature independent. The effect of annealing is found to be stable in time. Contrary to commonly observed behaviour of the intrinsic electrooptic coefficients, m_{22} changes significantly with temperature. In our opinion it would be of interest to know the temperature dependence of ε_2 for various x_c and consider the relationship between x_c and the temperature independent contribution to ε_2.

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