Phase matching for second harmonic generation in KNbO$_3$:Ta crystals with Nd:YAG- and GaAs-laser wavelengths

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Noncritical phase-matched second harmonic generation was measured in optically biaxial KNb$_{1-x}$Ta$_x$O$_3$ mixed crystals in the orthorhombic phase. For Nd:YAG-(1064 nm) the coefficient $d_{31}$ of the tensor of the nonlinear susceptibility was applied, while for the GaAs-laser (905 nm) the coefficient $d_{32}$ was used. For both laser wavelengths the phase-matching temperature decreases with increasing Ta concentration. Noncritical phase-matching at room temperature can be reached with the GaAs-laser for a Ta concentration of $\approx 9\%$. The corresponding value for the Nd:YAG laser is $\approx 14\%$.

INTRODUCTION

For pure KNbO$_3$ crystals efficient second harmonic generation (SHG) of near infrared laser light by noncritical phase matching has been reported recently.$^{1,2}$ The phase-matching temperature depends on the laser wavelength and the nonlinear optical coefficient used thus varying from $-36^\circ C$ up to $188^\circ C$. The dispersion [Fig. 1(a)] and the temperature dependence of the refractive indices [Fig. 1(b)] of KNbO$_3$ elucidate that noncritical phase matching can be reached for Nd:YAG-laser (1064 nm) by applying the $d_{31}$ coefficient of the tensor of the nonlinear susceptibility, while for the GaAs laser (905 nm) noncritical phase-matched SHG is possible using the $d_{32}$ coefficient.

Measurements of the dielectric constant$^3$ show in accordance with Triebwasser$^4$ that crystals grown from mixtures of ferroelectric KNbO$_3$ and the incipient ferroelectric KTaO$_3$ have decreasing phase transition temperatures of all three phase transitions (cubic$\rightarrow$tetragonal$\rightarrow$orthorhombic$\rightarrow$rhombohedral) with increasing Ta content (Fig. 2). Accordingly, the refractive indices are slightly varied, thus also varying the phase-matching temperatures of noncritical SHG. In the orthorhombic phase KNb$_{1-x}$Ta$_x$O$_3$ is an optically biaxial crystal, where the refractive indices obey the inequality $n_x < n_y < n_b$. Thus different geometries and fundamental laser wavelengths may be applied for noncritical phase-matched SHG, using the nonlinear coefficients $d_{11}$ and $d_{32}$. Since KNb$_{1-x}$Ta$_x$O$_3$ crystals can be grown over the whole concentration range it is of interest to study the variation of the SHG phase-matching conditions with composition.

EXPERIMENT

Our SHG experiments were performed on KNb$_{1-x}$Ta$_x$O$_3$ crystals grown from a K$_2$O-rich solution. The Ta-concentration in the samples was 0.0, 0.54, 2.4, 4.7, and 10.3 mol\% Ta as measured by electron microprobe analysis.$^{3,6}$ The phase-matching conditions were investigated for Nd:YAG- (1064 nm) and GaAs-laser light (905 nm). Two different experimental setups were used:

With the Nd:YAG-laser spatially resolved SHG measurements (SRSHG)$^{5,7}$ were performed. As fundamental light source served the expanded beam of a Nd:YAG-laser with a peak power of $\approx 10$ kW and a repetition rate of 1 kHz. The second harmonic light was detected by an optical multichannel analyzer (OMA) after appropriate filtering [Fig. 3(a)]. Thus both sample homogeneity and phase-matching temperature could be measured in one experiment.

The GaAs-laser—because of its lower power ($\approx 1$ W peak power and 5 kHz repetition rate)—had to be focused to the sample [Fig. 3(b)]. Beam divergence and spectral distribution were reduced by spatial and spectral filtering. The generated SHG-signal was detected by a photomultiplier and standard boxcar techniques.

The sample temperature in both setups could be varied by computer controlled peltier cooling and heating.

RESULTS AND DISCUSSION

The phase-matching conditions were investigated in the orthorhombic phase of the optically biaxial KNb$_{1-x}$Ta$_x$O$_3$ crystals, where the refractive indices obey the inequality $n_x < n_y < n_b$. For the Nd:YAG-laser wavelength (1064 nm) a geometry using the $d_{31}$ coefficient was applied, for the GaAs laser (905 nm) the $d_{32}$ coefficient was used. A contour plot (Fig. 4) of the sample with a Ta concentration of 10.3% (phase-matching temperature of $\approx 71^\circ C$) illustrates the very good homogeneity of the investigated samples.

The experimental results for the phase-matching temperatures are shown in Fig. 5. The temperatures for noncritical phase matching are strongly decreasing with increasing Ta concentration for both laser wavelengths. Noncritical phase matching in the orthorhombic phase is possible for all investigated crystals. The relation between the phase-matching temperature and the Ta concentration is nearly linear for both laser wavelengths with a negative slope of $\approx 11.5^\circ C$/mol\% Ta. Thus for the GaAs-laser noncritical phase matching at room temperature can be reached for a Ta concentration of $\approx 9\%$, the corresponding extrapolated value for the Nd:YAG-laser is $\approx 14\%$.

The SRSHG measurements show that the variation of the phase-matching temperature over the whole crystal is...
less than 1.5 °C (Fig. 4). With the linear relation of Fig. 5 a maximum variation of \( \approx 0.13\% \) of the Ta concentration can be estimated.

Because of the measured strong dependence of the phase-matching temperature from the Ta concentration, SRSHG measurements\(^6,^9\) can be used as an effective probe for characterizing \( \text{K}_{\text{Nb}_1-x}\text{Ta}_x\text{O}_3 \). From the phase-matching temperature the Ta content can be derived, the variation of the phase-matching temperature yields the sample homogeneity. If one assumes an accuracy of 0.3 °C of the determination of the phase-matching temperature, by SRSHG variations of the Ta concentration down to \( \approx 0.03 \text{ mol}\% \) Ta can be detected with a spatial resolution of \( \approx 20 \mu\text{m} \). Because of the assumed wide hysteresis

\( \approx 50 \degree \text{C} \) for \( \text{K}_{\text{Nb}_1-x}\text{Ta}_x\text{O}_3 \), this criterion for homogeneity is restricted to the concentration range of 0.0–15.5 mol% Ta.

If the SHG-intensity measured at a certain crystal position is plotted versus temperature \( T \) a sinc\(^2\)-like dependence is to be expected. The product of the half-width \( \Delta T \) and the crystal length \( L \) in the beam direction of the fundamental and the second harmonic wave can be used as a criterion for homogeneity\(^10\) in the beam direction. All investigated crystals show half-widths in the same order of magnitude as for pure \( \text{K}_{\text{Nb}_1-x}\text{Ta}_x\text{O}_3 \) \( \Delta T \cdot L \approx 0.29 \degree \text{C} \cdot \text{cm} \) indicating good homogeneity also in this direction.

In spite of the very good homogeneity of all samples investigated it was difficult to get crystals completely monodomain. This could be due to internal stress, point defects
FIG. 5. Temperatures for noncritically phase-matched SHG vs Ta concentration (squares) for Nd:YAG- and GaAs-laser. Circles and triangles indicate the orthorhombic—tetragonal phase transition and the orthorhombic—rhombohedral phase transition, respectively (calculated from Ref. 4).

and dislocations blocking the alignment of ferroelectric domains. Further efforts in improving the crystal growing techniques are needed to optimize the crystal quality. The above results show that KNb1-\(x\)Ta\(_x\)O\(_3\) with appropriately chosen \(x\) is a suitable material for room temperature SHG applications using near infrared lasers.

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