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 $KNb_{1-x}Ta_xO_3$ 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 °C up to 188 °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↔tetragonal ↔orthorhombic↔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 $KNb_{1-x}Ta_xO_3$ is an optically biaxial crystal, where the refractive indices obey the inequality $n_c < n_a < n_b$. Thus different geometries and fundamental laser wavelengths may be applied for noncritical phase-matched SHG, using the nonlinear coefficients d_{31} and d_{32}. Since $KNb_{1-x}Ta_xO_3$ 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 $KNb_{1-x}Ta_xO_3$ crystals grown from a K_2O-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)[8,9] were performed. As fundamental light source served the expanded beam of a Nd:YAG-laser with a peak power of $\sim$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 $KNb_{1-x}Ta_xO_3$ crystals, where the refractive indices obey the inequality $n_c < n_a < n_b$. 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 °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 °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

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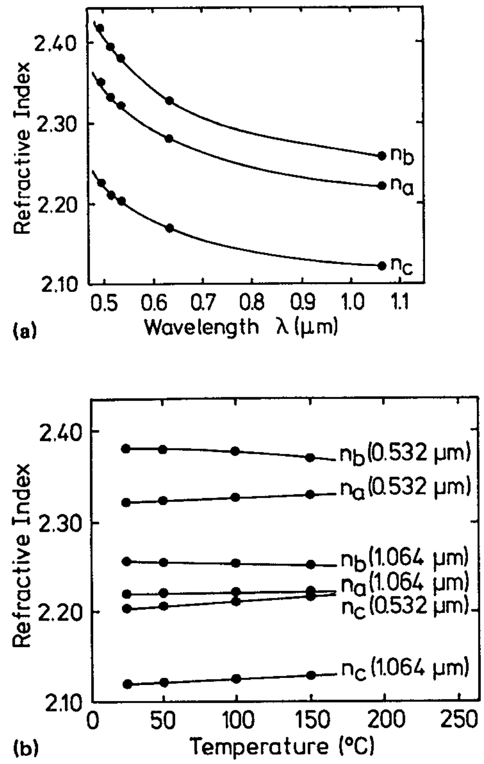

FIG. 1. (a) Dispersion of $KNbO_3$ at room temperature. (b) Temperature dependence of the refractive indices of $KNbO_3$ at 532 and 1064 nm.[5]

less than 1.5 °C (Fig. 4). With the linear relation of Fig. 5 a maximum variation of ≈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[8,9] can be used as an effective probe for characterizing $KNb_{1-x}Ta_xO_3$. 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 ≈0.03 mol% Ta can be detected with a spatial resolution of ≈20 μm. Because of the assumed wide hysteresis (≈50 °C for $KNbO_3$[7]) of the orthorhombic ↔rhombohedral phase transition of $KNb_{1-x}Ta_xO_3$ 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 ΔT 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 $KNbO_3$ ($\Delta T \cdot L \approx 0.29$ °C cm)[5] 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

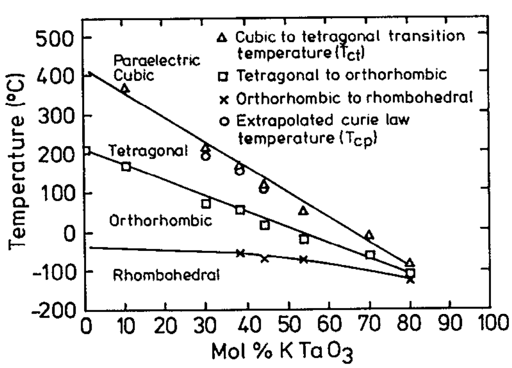

FIG. 2. Dependence of the phase transition temperatures of $KNb_{1-x}Ta_xO_3$ from the Ta concentration.[4]

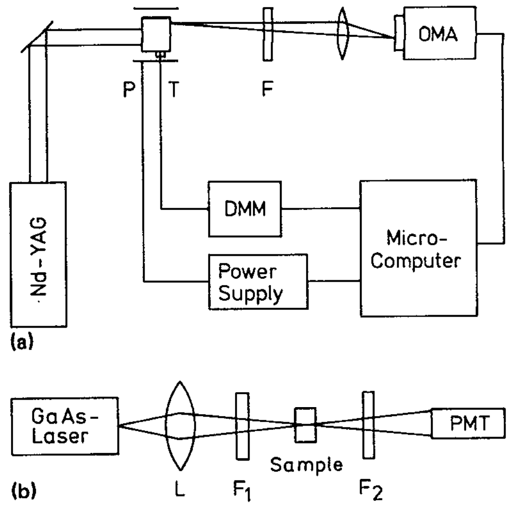

FIG. 3. (a) Experimental setup for spatially resolved SHG with Nd:YAG-laser (OMA: optical multichannel analyzer, P: peltier-cooling/heating, T: thermocouple, F: short-pass filter BG 18). (b) Setup for phase-matched SHG with GaAs-laser (L: focusing lens, F1: interference filter for 905 nm, F2: short-pass filter BG 18).

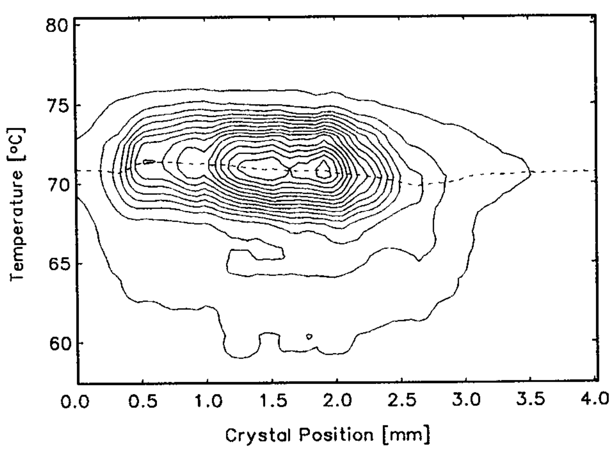

FIG. 4. Contours of constant intensity of the spatially resolved SHG measurement of $KNb_{1-x}Ta_xO_3$ with a Ta concentration of 10.3%. The dashed line shows the phase-matching temperature versus crystal positon.

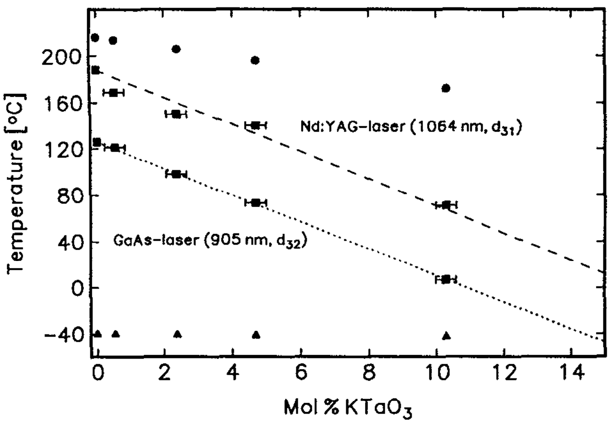

FIG. 5. Temperatures for noncritically phase-matched SHG vs Ta- concentration (squares) for Nd:YAG- and GaAs-laser. Circles and triangles indicate the orthorhombic→tetragonal phase transition and the orthorhombic→rhombohedral phase transition, respectively (calculated from Ref. 4).

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 $KNb_{1-x}Ta_xO_3$ with appropriately chosen x is a suitable material for room temperature SHG applications using near infrared lasers.

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