Radiation Effects and Defects in Solids 119-121:281 (1991)

FREQUENCY DOUBLING OF Nd:YAG- AND GaAs-LASERS BY $KNbO_3$: Ta CRYSTALS

N. SCHMIDT, P. VOIGT, B. HELLERMANN, K. BETZLER, H. HESSE, AND S. KAPPHAN
Fachbereich Physik, Universität Osnabrück, FRG

Abstract For $KNb_{1-x}Ta_xO_3$ crystals the influence of the Ta-concentration on the phase-matching properties for optical second harmonic generation (SHG) was measured. For non-critical phase matched SHG of the Nd:YAG-laser (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. Non-critical 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

The maximum storage capacity of optical storage media (CD-ROM, WORM etc.) is inversely proportional to the square of the laser wavelength used. Thus it is highly desirable to use short wavelength light sources. As semiconductor lasers, the commonly used sources, are not available for short wavelengths, optical frequency doubling of near infrared lasers is one way to overcome this limitation.

For pure $KNbO_3$ crystals efficient second harmonic generation (SHG) of near infrared laser light by non-critical 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 (figure 1) and the temperature dependence of the refractive indices (figure 2) of $KNbO_3$ elucidate that non-critical 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-

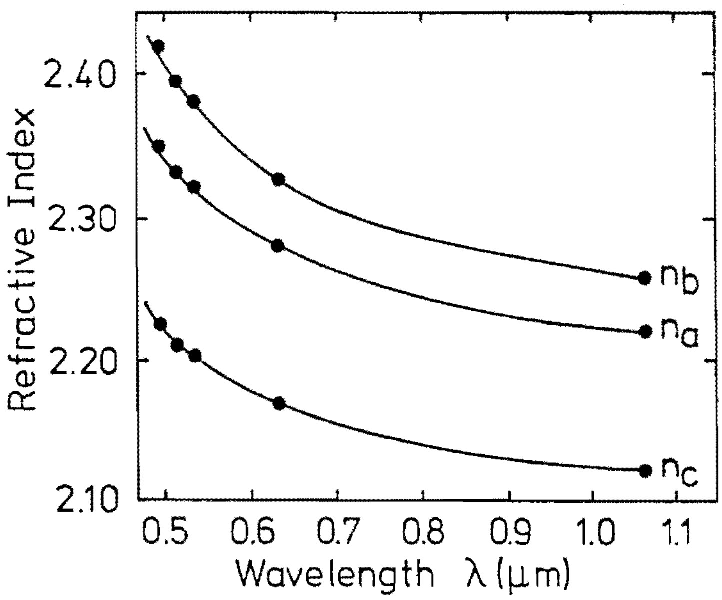

Figure 1: Dispersion of $KNbO_3$ at room temperature (Uematsu [5]).

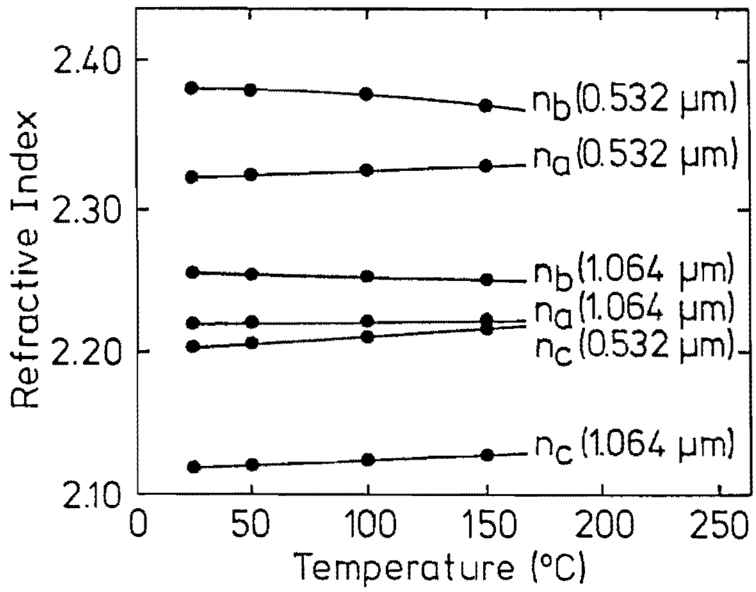

Figure 2: Temperature dependence of the refractive indices of $KNbO_3$ at 532 nm and 1064 nm (Uematsu [5]).

laser (905 nm) non-critical phase-matched SHG is possible using the d_{32} coefficient.

Measurements of the dielectric constant [3] show in accordance with

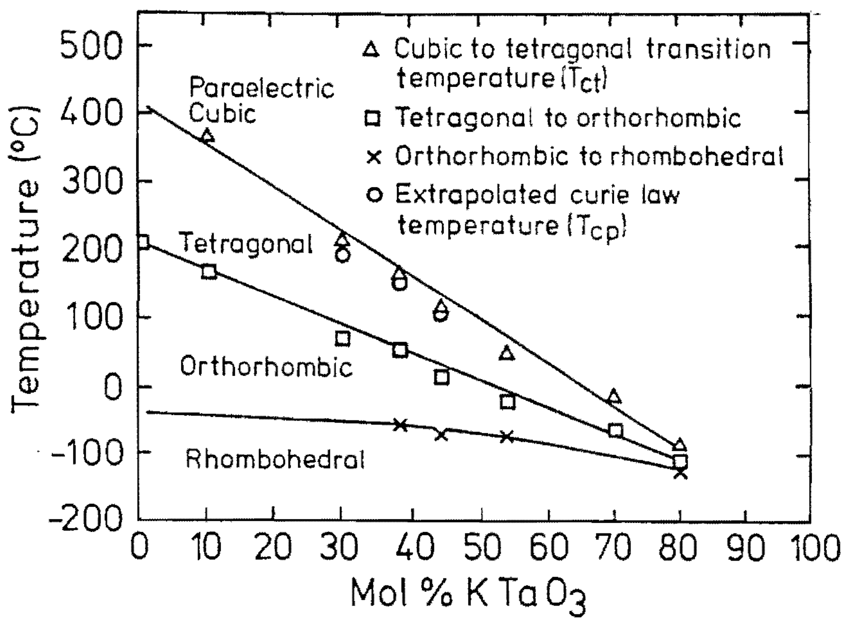

Figure 3: Dependence of the phase transition temperatures of $KNb_{1-x}Ta_xO_3$ from the Ta-concentration [4].

Triebwasser [4] that the partial replacement of Nb by Ta in $KNbO_3$ decreases the phase transition temperatures of all three phase transitions (cubic ↔ tetragonal ↔ orthorhombic ↔ rhombohedral) with increasing Ta-content (figure 3). Accordingly, the refractive indices are slightly varied, thus also varying the phase-matching temperatures of non-critical 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 non-critical 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.

EXPERIMENTAL

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).

The Nd:YAG-laser used had a peak power of ≈ 10 kW and a repetition rate of 1 kHz, the respective values for the GaAs-laser were 1 W and 5 kHz.

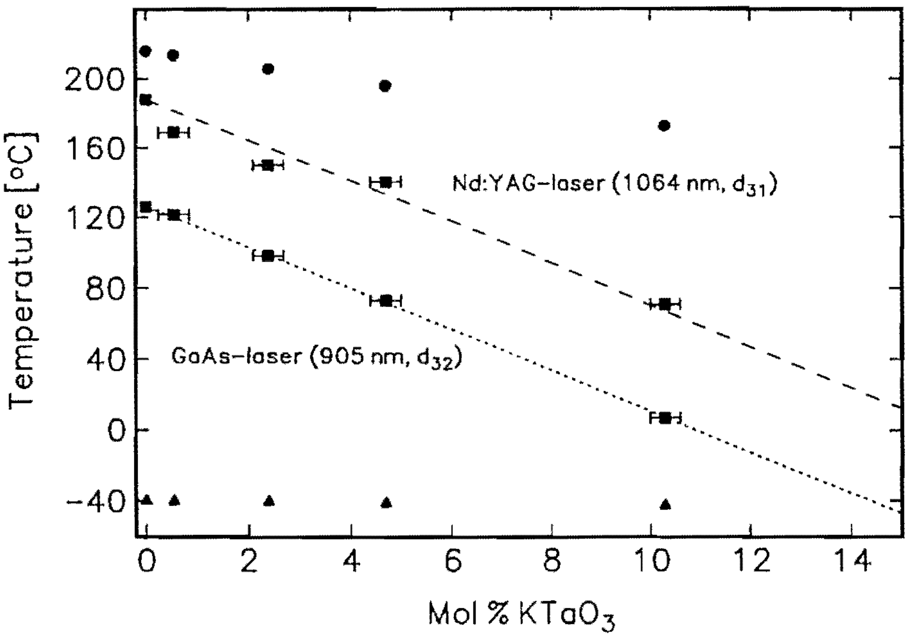

Figure 4: Temperatures for non-critically phase-matched SHG versus Ta-concentration (squares) for Nd:YAG- and GaAs-laser. Circles and triangles indicate the orthorhombic → tetragonal phase transition and orthorhombic → rhombohedral phase transition (calculated according to ref. 4).

The beam divergence and spectral width of the GaAs-laser were reduced by spatial and spectral filtering. The generated SHG-signal was detected by a photomultiplier and standard boxcar techniques.

The sample temperature 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 (1064 nm) a geometry using the d_{31} coefficient was applied, for the GaAs-laser (905 nm) the d_{32} coefficient was used.

The experimental results for the phase-matching temperatures are shown in figure 4. The temperatures for non-critical phase-matched SHG are strongly decreasing with increasing Ta-concentration for both laser wavelengths. Non-critical phase-matching in the orthorhombic phase is possible for all investigated crystals. The relation between the phase-matching

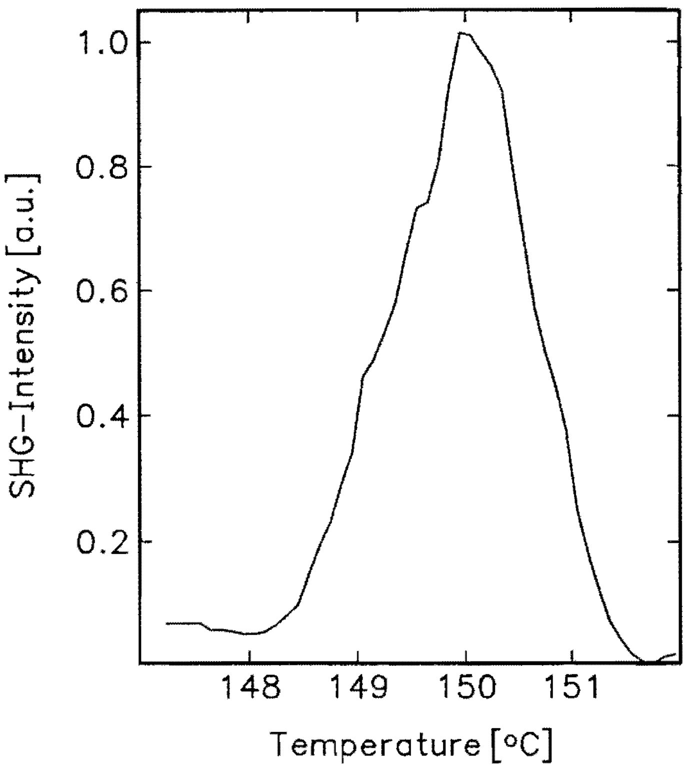

Figure 5: The temperature dependence of the SHG-intensity for the $KNb_{1-x}Ta_xO_3$ crystal with a Ta-concentration of 2.39 mol %. The sample length was ≈ 2 mm yielding a halfwidth of $\Delta T \cdot L \approx 0.25\,°C \cdot cm$.

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 non-critical 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 $\approx 14\,\%$.

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 [9] in the beam direction. All investigated crystals show half-widths in the same order of magnitude as for pure $KNbO_3$ ($\Delta T \cdot L \approx 0.29\,°C \cdot cm$) [5] indicating good homogeneity in this direction (figure 5).

Because of the measured strong dependence of the phase-matching temperature from the Ta-concentration, spatially resolved SHG measurements (SRSHG) [7,8] should be an effective tool to probe the homogeneity of

$KNb_{1-x}Ta_xO_3$ crystals. Measurements using the SRSHG technique are in progress.

In the present experiments it was difficult to get crystals completely monodomain. This could be due to internal stress, point defects and dislocations blocking the alignment of ferroelectric domains [10]. Further investigations to clarify this situation are in progress. Anyway 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.

Acknowledgements We thank B. Gather for measuring the Ta-concentration with microprobe techniques of some of our samples used. The work was supported by the Deutsche Forschungsgemeinschaft in the framework of SFB 225.

References

[1] Baumert, J. C., Hoffnagle, J., Günter, P. (1984). Nonlinear optical effects in $KNbO_3$ crystals at $Al_xGa_{1-x}As$, dye, ruby and Nd:YAG laser wavelengths. *SPIE-Proceedings (ECOOSA 84, Amsterdam)*, 492

[2] Günter, P., Asbeck, P. M., Kurtz, S. K. (1979). Second harmonic generation with $Al_xGa_{1-x}As$ lasers and $KNbO_3$ crystals. *Applied Physics Letters* **35**, 461 – 463

[3] Baller, F., Gather, B., Hellermann, B., Hesse, H., Krätzig, K. (1990). The influence of stress on the dielectric properties of KTN crystals. *Physica Status Solidi* **116**, K195 – K198

[4] Triebwasser, S. (1959). Study of ferroelectric transitions of solid-solution single crystals of $KNbO_3 - KTaO_3$. *Physical Review* **114**, 63 – 70

[5] Uematsu, Y. (1974). Nonlinear optical properties of $KNbO_3$ single crystals in the orthorhombic phase. *Japanese Journal of Applied Physics* **13**, 1362 – 1368

[6] Hellermann, B., Baller, F., Blachnik, R., Gather, B., Hesse, H., Schnitter, J. (1990). Crystal growth and characterisation of $K(Ta_{1-x}Nb_x)O_3$-solid solutions. *Thermochimica Acta* **160**, 71 – 77

[7] Schmidt, N., Betzler, K., Kapphan, S. (1987). Spatially resolved second harmonic generation and interferometric investigations of $LiNbO_3$ crystals. *Crystal Lattice Defects and Amorphous Materials* **15**, 103 – 108

[8] Schmidt, N., Betzler, K., Grabs, M., Kapphan, S., Klose, F. (989). Spatially resolved second harmonic generation investigations of proton induced refractive-index changes in $LiNbO_3$. *Journal of Applied Physics* **65**, 1253 – 1256

[9] Nash, F. R., Boyd, G. D., Sargent III, M., Bridenbaugh, P. M. (1970). Effect of optical inhomogenities on phase matching in nonlinear crystals. *Journal of Applied Physics* **41**, 2564 – 2576

[10] Betzler, K., Hellermann, B., Hesse, H. (1987). Influence of the dielectric constant anisotropy on the poling of ferroelectrics. *Ferroelectric Letters* **7**, 143 – 147