Study of KTiOPO₄ gray-tracking at 1064, 532, and 355 nm

B. Boulanger[a)] and M. M. Fejer

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

R. Blachman and P. F. Bordui

Crystal Technology Inc., Palo Alto, California 94303

Photochromic "gray-track" damage occurring during 1064 nm second-harmonic generation (SHG) in KTiOPO₄ is a significant limitation in many practical applications. Measurements of the evolution of gray-track damage during SHG, along with measurements of the 355 nm radiation simultaneously produced by non-phase-matched sum-frequency generation, are described. Comparison of these measurements with the gray-tracking induced by exposure to a single wavelength indicates that for the conditions investigated here, the gray-tracking during 1064 nm SHG is dependent only on the intensity of the 532 nm radiation. The dependence of the induced absorption on the 532 nm intensity is nonlinear, having a threshold of 80 MW/cm², and an approximately linear increase for intensities above this threshold. © *1994 American Institute of Physics.*

Photochromic damage (gray-tracking) often occurs in KTiOPO₄ (KTP) during high peak power nonlinear frequency conversion interactions. Different hypotheses have been advanced to explain this phenomenon which led to several prior studies. In 1986 Driscoll *et al.*[1] suggested that the gray-tracking during 1064 nm SHG is due to a two-photon process requiring both 1064 and 532 nm beams. In 1992 Bosenberg *et al.*[2] observed gray-tracks during OPO pumped at 532 nm and generating over 700–900 and 1300–2200 nm. In 1992 Loiacono *et al.*[3] suggested that the gray-track mechanism involves Ti³⁺; they showed that gray-tracks can be formed using only 532 nm. In 1993 Blachman *et al.*[4] observed UV generated during 1064 nm SHG and showed that a gray-track can be induced by 355 nm radiation alone. Tyminski[5] observed, in addition to the photochromic effect, an astigmatic beam distortion of the harmonic beam which can be interpreted as a photorefractive effect occurring with simultaneous infrared and green illumination in hydrothermally grown KTP.

In this work we determined the relative importance of both individual and simultaneous exposure to IR, visible, and UV radiation, and carried out initial studies of the time dependence of the darkening and recovery processes. The goal is to parameterize the gray-track process to enable quantitative device design, as well as to contribute to an understanding of the fundamentals of the gray-track phenomena in KTP. Crystals used in this work were produced by slow-cooled immersion-seeded solution growth from a K₆P₄O₁₃ flux.[6]

The first purpose of this work was to quantify the generation of 355 nm radiation and to determine its importance in gray-tracking during type-II 1064 nm SHG phase-matched in $x-y$ plane ($\theta=90°$, $\phi=23°$) of KTP. The SHG involves ordinary (o, polarized perpendicular to the z axis) and extraordinary (e, polarized orthogonally to o) waves at 1064 nm and an o wave at 532 nm. Only two configurations of polarizations have a nonzero effective coefficient d_{eff} for the indirect third-harmonic generation (THG) by summing 1064 and 532 nm: o waves at 355 and 532 nm and an e wave at 1064 nm, or o waves at 532 and 1064 nm and an e wave at 355 nm. The corresponding coherence lengths and figures of merit are $l_{c,o}=2.26$ μm and $(d_{\text{eff}}^2/n^3)_o=0.95$ pm/V, and $l_{c,e}=0.69$ μm and $(d_{\text{eff}}^2/n^3)_e=0.93$ pm/V, respectively. In these expressions n^3 represents the product of the refractive indices evaluated for the polarizations and frequencies of the three waves involved in the interaction. The refractive index data of Ref. 7 and nonlinear susceptibility data of Ref. 8 (extrapolated by Miller's rule) were used in evaluating these expressions.

The small coherence lengths for THG imply that the conversion to the third harmonic does not significantly deplete the fundamental or the second harmonic, and that the amplitudes of the fundamental and second harmonic do not change significantly over one coherence length. Under these assumptions, neglecting the walk-off and taking the fundamental input to be a Gaussian beam of $1/e^2$ radius w_o, the generated e-polarized third-harmonic power $P_{3\omega,e}(z)$ inside the crystal can be accurately approximated by

$$P_{3\omega,e}(z)=\Gamma_e P_\omega(z)P_{2\omega}(z)(l_{c,e}/w_o)^2, \tag{1}$$

where

$$\Gamma_e=\frac{8.55\times10^{-8}}{2\pi^2\lambda^2}\left(\frac{d_{\text{eff}}^2}{n^3}\right)_e \tag{2}$$

and P_ω and $P_{2\omega}$ are the powers inside the crystal at the fundamental and second harmonic, respectively, and λ the vacuum wavelength of the fundamental expressed in micrometers. For the o-polarized output, similar expressions hold, with e replaced by o. Numerically, we find $\Gamma_e=2.67\times10^{-9}$ W⁻¹, and $\Gamma_o=2.78\times10^{-9}$ W⁻¹. $P_{3\omega}(z)$ is independent of the absorption coefficient at 355 nm (≈0.98 cm⁻¹ for both o and e polarizations) because the absorption length is roughly $10^4\times$ higher than the coherence lengths.

[a)]Visiting Scientist, at the new address: Laboratoire de Physique du Solide, Unite de Recherche Associee 1796, Centre National de la Recherche Scientifique, Universite de Bourgogne, UFR des Sciences et Techniques, Batiment Mirande, BP 138 21004 Dijon, France.
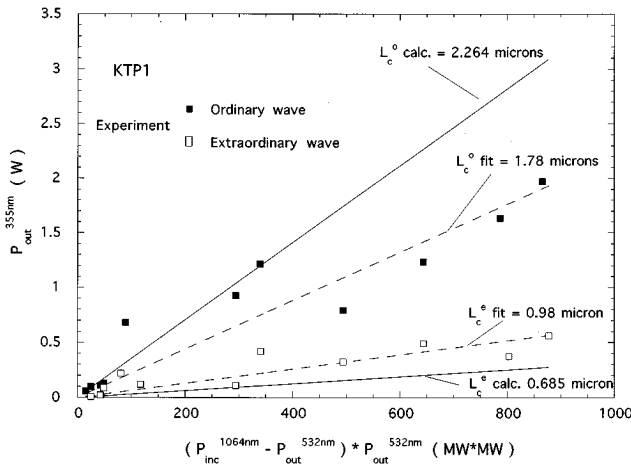
FIG. 1. Ordinary and extraordinary 355 nm peak powers generated by indirect non-phase-matched 1064 nm THG during type-II phase-matched 1064 nm SHG: propagation direction $\theta=90°$, $\phi=23.3°$, crystal length=1 cm. Lines are theoretical predictions and best linear fits to experimental data.

According to Eq. (1), the 355 nm generated by each coherence length reaches a maximum value when the product of the 1064 and 532 nm powers is maximum, that is to say, for a 50% SHG efficiency.

We performed a type-II SHG experiment with a 10 Hz, injection-seeded, single-longitudinal-mode, 1064 nm Nd:YAG (Spectra-Physics DCR-2A-10) laser equipped with super-Gaussian mirrors. The pulse is 10 ns in duration (FWHM) and is near a Gaussian single-transverse mode. The beam diameter is 4 mm in radius and polarized at 45° to the principal axes of the 10 mm long KTP crystal. The incident fundamental and generated second- and third-harmonic energies were simultaneously measured; the 355 nm energy was measured with a boxcar integrator and a silicon photodiode previously calibrated at 355 nm against a pyroelectric joulemeter; the 1064 and 532 nm energies were measured directly with two joulemeters. A series of dichroic mirrors, prisms, and filters were used to separate the beams. A Glan polarizer allowed the measurement of the two 355 nm polarization components. The incident fundamental energy could be controlled using a rotating half-wave plate and a Glan polarizer placed at the exit of the laser.

The SHG experiment was performed with fundamental energies between 78 mJ (62 MW/cm^2) and 590 mJ (470 MW/cm^2). For each experiment the crystal was rotated in ϕ to obtain the maximum SHG output. The peak SHG power conversion efficiency, estimated from the measured energy conversion efficiency multiplied by the ratio between the fundamental and harmonic pulse durations ($\tau_\omega/\tau_{2\omega}=\sqrt{2}$), increased from 50% at 63 MW/cm^2 to a maximum value of 85% at 200 MW/cm^2 and decreased for higher intensities, reaching 50% at 470 MW/cm^2; this typical behavior corresponds to the "overdriving" of the crystal, where efficiency decreases with increased fundamental intensities.[9]

Two orthogonally polarized components at 355 nm were detected. The powers are plotted in Fig. 1 as a function of the product of the 532 and 1064 nm powers at the exit of the crystal, along with the theoretical lines according to Eq. (1),

corrected by the product of Fresnel transmission coefficients of the three waves, and Eq. (2), and best linear fits. The ratio of the theoretical and experimental slopes is 1.62 and 0.49 for the o and e waves, respectively, suggesting that the generated UV is adequately modeled as that produced in a single coherence length of the crystal. The small discrepancies are probably due to errors in the refractive index data and nonlinear susceptibilities near the UV edge of KTP. We were unable to reproduce the large intensities of UV radiation that have been reported anecdotally and had previously been observed qualitatively.[4]

Two kinds of damage were observed during these experiments: KTP began to gray-track for a pump intensity of about 90 MW/cm^2 (corresponding to a peak power SHG efficiency of 60%). Above this intensity, the crystal became progressively darker until 480 MW/cm^2; at this intensity the exit face was damaged to a depth of 0.3 mm over a 2 mm diameter area. In all cases, the gray-track totally disappeared within 24 h following the experiment.

Having established typical intensities at the three wavelengths during gray-track generation, we then investigated the relative importance of the contributions of the 1064, 532, and 355 nm radiation. It is not possible to study these frequency components individually for radiation propagating in a phase-matched direction as o-532 nm input generates o- and e-1064 nm by parametric fluorescence, and o- and e-355 nm by summing the pump and the fluorescence radiation. Thus we studied three presumably identical 15 mm long crystals, designated KTP1, KTP2, and KTP3, with propagation along a direction in the $x-y$ plane at $\theta=47.5°$, an arbitrary direction chosen far from the 1064 nm SHG phase-matching direction at $\phi=23.3°$ in the $x-y$ plane. We chose to quantify the darkening by measuring the 532 nm transmission coefficient.

Two consecutive experiments were performed on KTP1. At first, KTP1 was illuminated by 532 nm only, with intensities between 65 and 190 MW/cm^2, neither 355 nor 1064 nm were detected during any of these exposures. The steady-state transmission coefficient T was measured with the 532 nm beam itself. T decreased monotonically with intensity. A measurement of T at 40 MW/cm^2 after each measurement at higher intensity led to the same value for T, indicating that nonlinear absorption of the 532 nm radiation was not significantly skewing the measurement of the absorption at the higher intensities. We show in Fig. 2 the steady-state absorption coefficient $\alpha=L^{-1}\ln(T/T_f)$, where $L=1.5$ cm is the crystal length and $T_f=0.846$ is the nominal Fresnel transmission of a lossless crystal. This experiment proves that 532 nm can alone gray-track KTP; it corroborates the work of Loiacono et al.[3] After 1 day KTP1 had recovered its initial transparency, similar to the recovery from UV induced gray-tracking observed previously.[4]

We repeated these measurements on KTP1, but with o-polarized 532 nm combined with 1064 nm e and o waves, using intensities similar to those encountered in typical SHG experiments where gray-tracking occurs. 355 nm e and o waves were detected with powers close to the values previously measured during SHG because the associated coherence lengths and figures of merit are close to those in the
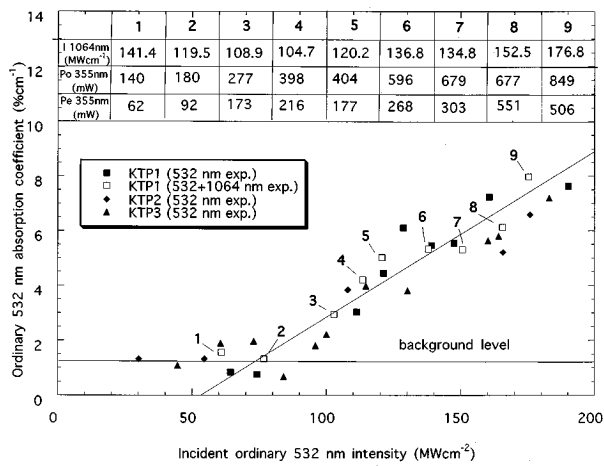
Boulanger et al.

FIG. 2. Comparison of the absorption induced in several nominally identical crystals from the same source, either with 532 nm exposure alone or with simultaneous 532 and 1064 nm exposure where 355 nm is generated. Propagation direction $\theta=47.5°$, $\phi=0°$, crystal length=1.5 cm. The continuous lines are guides for the eyes. The look-up table concerns both 532 and 1064 nm exposure of KTP1.

FIG. 3. Time evolution of gray-tracking for two different incident 532 nm intensities, 180 and 110 MW/cm². Propagation direction $\theta=47.5°$, $\phi=0°$, crystal length=1.5 cm. The continuous lines are exponential fits to the experimental data.

SHG phase-matching direction: $l_{c,o}=1.87$ μm, $(d_{\mathrm{eff}}^2/n^3)_o=1.05$ pm²/V² and $l_{c,e}=1.02$ μm, $(d_{\mathrm{eff}}^2/n^3)_e=1.22$ pm²/V². The 532 nm absorption coefficient and the associated 355 nm powers and 1064 nm intensities are given in Fig. 2; it shows the same behavior as with 532 nm exposure only. Thus we can conclude that the intensity of the incident 1064 and generated 355 nm present during SHG are not sufficient to affect the gray-tracking induced by the 532 nm radiation. Under the conditions of these experiments, the intensity of the 532 nm radiation alone determines the gray-track absorption in typical SHG configurations.

We also show in Fig. 2 the absorption coefficient measured during a 532 nm exposure of KTP2 and KTP3. All the values are close to those of KTP1. These experiments indicate clearly that the gray-tracking effect has a 532 nm intensity threshold of about 80 MW/cm² for these crystals at a 10 Hz Q-switch frequency. This threshold is similar to that observed during our SHG experiments (108 MW/cm² of 532 nm) and with the OPO experiments reported in Ref. 2 (80 MW/cm²). Above the threshold, the absorption increases approximately linearly with a slope of about 6.5×10^{-4} cm²/MW. The extrapolation of this straight line does not pass through the origin, which indicates a nonlinear mechanism for the gray-tracking process, consistent with the UV damage measurements of Ref. 4.

We also measured the time evolution of the absorption in KTP2 with 532 nm exposure only, shown for two gray-tracking experiments with 107 and 176 MW/cm² in Fig. 3. The absorption exhibited saturating exponential behavior, with time constants of about 230 and 290 s, respectively. After termination of the exposure the absorption decayed back to the original background level, over times much longer than those required to create the gray-track. The decay was approximately linear, with a rate of 0.75%/h for both exposure levels (Fig. 4). These observations are not consistent with a two-photon absorption mechanism for the creation of the absorbing centers if a simple thermal decay
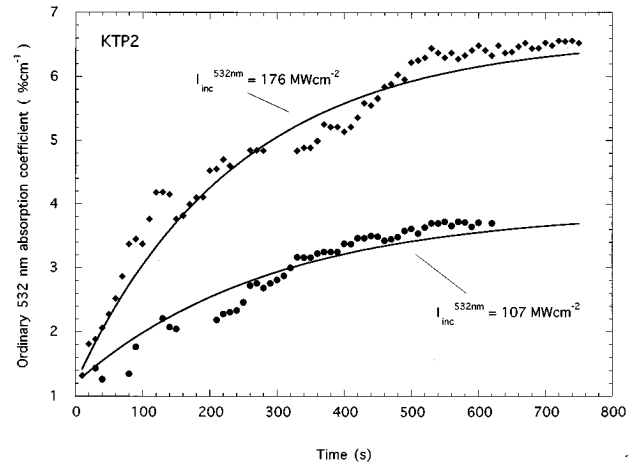
mechanism is postulated for establishing the steady state. Significant photobleaching of the absorbing centers themselves by the 532 nm radiation could explain much of the deviation from this simple model. To test this possibility, we exposed a sample, gray-tracked with 100 MW/cm², to a 532 nm intensity of 55 MW/cm² (just below the "gray-track threshold"), and recorded the decay of the absorption. No resolvable difference between the decay under laser illumination and the room light decay was observed. Thus the underlying mechanisms and kinetics of decay are not well understood.

We have clearly established that in these experiments, the gray-tracking effect during 1064 nm SHG in KTP was due to a nonlinear process involving only the 532 nm radiation, with no influence of either 1064 or 355 nm radiation on the darkening at intensities typical of SHG interactions. We have also demonstrated that the gray-tracking has a threshold of about 80 MW/cm² at 532 nm for 10 Hz, 10 ns pulses in the crystals studied. All observed gray-tracks were reversible.

[1] T. A. Dricoll, H. J. Hoffman, R. E. Stone, and P. E. Perkins, J. Opt. Soc. Am. B **3**, 683 (1986).

[2] W. R. Bosenberg and D. R. Guyer, Appl. Phys. Lett. **61**, 387 (1992).

[3] G. M. Loiacono, D. N. Loiacono, T. McGee, and M. Babb, J. Appl. Phys. **72**, 2705 (1992).

[4] R. Blachman, P. F. Bordui, and M. M. Fejer, Appl. Phys. Lett. **64**, 1318 (1994).

[5] J. K. Tyminski, J. Appl. Phys. **70**, 5570 (1991).

[6] P. F. Bordui, J. C. Jacco, G. M. Loiacono, R. A. Stolzenberger, and J. Zola, J. Cryst. Growth **84**, 403 (1987).

[7] K. Kato, IEEE J. Quantum Electron. **QE-27**, 1137 (1991).

[8] B. Boulanger, J. P. Feve, G. Marnier, B. Menaert, X. Cabirol, P. Villeval, and C. Bonnin, J. Opt. Soc. Am. B **11**, 750 (1994).

[9] D. Eimerl, IEEE J. Quantum Electron. **QE-23**, 575 (1987).