Repetition rate dependence of gray-tracking in $KTiOPO_4$ during second-harmonic generation at 532 nm

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The aim of this work is to study gray tracking during type II second-harmonic-generation 1064→532 nm in halide flux grown $KTiOPO_4$ crystals. The repetition rate of the laser varies from 1 to 20 kHz. We experimentally demonstrate that the gray-tracking threshold, expressed by the harmonic peak power density, is a decreasing exponential function of the Q-switch frequency. The corresponding 532 nm average power density, however, remains constant. [S0003-6951(97)00603-7]

Photochromic damage occurs during high power nonlinear optical frequency conversion interactions. Gray tracking is reported during long term experiments, particularly for 1064 nm second-harmonic generation (SHG), where the darkening of the $KTiOPO_4$ (KTP) crystal is observed;[1–3] this can lead to a substantial reduction of the SHG conversion efficiency. A probe beam is used to see the simultaneous increase of absorption.[2]

Under usual 1064 nm SHG experimental conditions, the gray-track formation is only caused by the 532 nm beam.[4,5] This corroborates the observation of similar gray tracks in a 532 nm pumped optical parametric oscillator (OPO) emitting an infrared beam.[6] Nevertheless, gray tracking can occur at other wavelengths: it is reported for 1047 nm SHG of Nd:YLF laser,[7] and under illumination at 355 nm.[8]

In addition, an astigmatic distortion of the harmonic beam occurs, especially under strong focusing; it may be interpreted in terms of photorefractive effects.[3,7,8]

A gray-tracking threshold of 80 MW/cm^2 at 10 Hz is determined by measurement of the absorption of 532 nm during irradiation.[5]

For the most part, previous studies have used Q-switched lasers with a pulse duration of about 10 ns, a repetition rate between 10 and 20 Hz, and a number of shots between 10^3 and 10^5. One is made at a higher frequency (4 kHz),[7] and one concerns the pico-second range.[2] This letter deals with the gray-tracking threshold evolution with respect to the repetition rate of the laser.

We study type II 1064→532 nm SHG for a propagation in the x-y plane of KTP ($\theta=90°$; $\phi=23.1°$) which corresponds to the maximal conversion efficiency.

The crystals are grown from halide flux,[9] with lengths between 1.16 and 7.5 mm. The 1064 nm pump laser is a Microcontrole Mod. 904 Nd:YAG which emits an unpolarized, one-time-diffraction-limited TEM_{00} Gaussian beam, longitudinally multimode. The 1064 nm beam propagates through a Glan polarizer and a half wave plate and is then focused in the studied KTP crystal with a 50 mm focal length lens. The variation of the generated harmonic power is obtained by the modification of the focusing conditions, by the rotation of the half wave plate with respect to the crystal axes, or by the use of different crystal lengths. The harmonic beam waist, $w_0^{2\omega}=w_0^{\omega}/\sqrt{2}$, is determined by the prospection method.[10] The laser is Q switched, with a repetition rate in the range 1–20 kHz. The pulse shape depends on the Q-switch frequency and it cannot be adjusted. The pulse duration is measured for both fundamental and harmonic beams: it has a Gaussian leading edge and a Lorentzian trailing edge; the full width at half maximum (FWHM) of the fundamental pulse varies from $\tau_\omega=104$ ns to $\tau_\omega=525$ ns for the considered range of repetition rate. The corresponding harmonic FWHM $\tau_{2\omega}$ range is 74–338 ns.

The generated harmonic beam is separated from the nonconverted fundamental by a prism. The average power conversion efficiency η_{average} is measured with two powermeters. The corresponding peak power conversion efficiency, deduced by the relation $\eta_{\text{peak}}=\eta_{\text{average}}\tau_\omega/\tau_{2\omega}$, varies between 5% and 62%.

For different Q-switch frequencies, several tests are performed for about 90 min each, which represent a number of shots between 4.10^6 and 10^8, according to the repetition rate. The SHG conversion efficiency and the transversal profile of the harmonic beam at the exit of the crystal are checked during each experiment. The crystal is observed through an X5 magnifying telescope after the illumination. For these tests, we distinguish three typical behaviors: (i) no damage appears, there is a constant conversion efficiency, there is no transversal deformation of the harmonic beam, and no visible damage inside the crystal; (ii) a slight decrease of the SHG efficiency, a slow and not clear deformation of the beam, and a very thin dark mark in the bulk of the crystal; (iii) clear and rapid damage formation: a strong reduction of the conversion efficiency, an astigmatic deformation of the harmonic beam, and a distinct dark track inside the KTP crystal.

All the observed gray tracks have a very long recovery time, of about one year or more, which is characteristic of strong focusing;[2,7] thus it is not yet possible to comment on the reversibility of the created gray tracks. On the contrary, a recovery time of about a day is observed for weak focusing experiments.[5,6,8]

The results concerning two particular Q-switch repetition rates, $f=1.060$ kHz for experiments 1–6, and f

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TABLE I. Considered situations for gray-tracking studies in KTP during type II SHG at 1064 nm: Q-switch frequency $f=1.060$ kHz (experiments 1–6) and $f=1.800$ kHz (experiments 7 and 8); fundamental beam waist radius $w_0^\omega=22$ μm (1–6), $w_0^\omega=40$ μm (7), and $w_0^\omega=20$ μm (8); pulse durations (FWHM) $\tau_\omega=104$ ns (1–6), $\tau_\omega=115$ ns (7 and 8), $\tau_{2\omega}=74$ ns (1–6), and $\tau_{2\omega}=75$ ns (7 and 8). Behaviors (i), (ii), and (iii) refer, respectively, to: no damage, estimated threshold, and strong damage. The terms $u_{\omega,2\omega}$ are the pulse energies and $I_{\omega,2\omega}$ the peak power densities of the fundamental and harmonic beams.

No.	Crystal length (mm)	u_ω (μJ)	I_ω (MW/cm^2)	$u_{2\omega}$ (μJ)	$I_{2\omega}$ (MW/cm^2)	Damage behavior	η_{peak} decreasing over 1 h
1	4.91	398	181	13.4	18.0	(i)	None
2	5.50	415	189	81.8	110	(ii)	None
3	5.50	415	189	101	136	(ii)	−3%
4	4.91	424	193	119	160	(iii)	−12%
5	4.91	419	190	126	169	(iii)	−31%
6	4.91	424	193	148	199	(iii)	−31%
7	4.90	388	51.7	122	48.8	(i)	None
8	4.90	352	187	128	205	(iii)	−13%

$=1.800$ kHz for tests 7 and 8, are given in Table I. The crystals used for these experiments were antireflection coated for both fundamental and harmonic wavelengths. The results confirm the respective importance of the fundamental and harmonic beams for the gray tracking;[4,5] for similar 1064 nm peak power densities, the observed damages totally differ, depending on the peak power density at 532 nm (experiments 1, 2, and 4), which corroborates the results obtained by one of us at 10 Hz.[5] The present experiments also show that the harmonic average power is not the relevant parameter: the same pulse energy at 532 nm can cause very different damages depending on the corresponding peak power density (experiments 7 and 8). As a first step we can define the gray-tracking threshold by the harmonic peak power density.

A quantitative determination of the threshold should require a very careful measurement of the optical absorption as a function of the 532 nm peak power density during SHG. However, for very strong focusing situations, the exact superposition of a probe beam with the very narrow illuminated volume of the crystal is difficult. The direct measurement of the absorption with the generated 532 nm radiation requires a precision that could not be achieved with our experimental setup. The thresholds we determine are thus only qualitative and the behavior (ii) represents the "threshold situation." Nevertheless, it is fully representative of the evolution of the phenomenon because we use the same visual criterion for all the experiments.

Figure 1 shows the observed damages and the corresponding harmonic peak power densities as a function of the repetition rate. An experiment which had been performed at 4 kHz[7] is also plotted on this graph: actually, for a 50 MW/cm^2 fundamental peak power density, the authors observe a slight degradation of the harmonic beam after 10^7 shots, which is typical of the threshold situation (ii); according to the reported efficiencies, we estimate the corresponding harmonic peak power density between 40 and 45 MW/cm^2. The values obtained in the present work correspond well with this result. Note that all these crystals are grown from a halide flux.[9] The threshold of 80 MW/cm^2 which had been measured at 10 Hz,[5] for crystals grown from a $K_6P_4O_{13}$ flux,[11] is lower than the value extrapolated from the present data at higher frequencies. It is not surprising because the value of 80 MW/cm^2 comes from precise measurements of the absorption at 532 nm under weak focusing, whereas the threshold criterion chosen in the present work is only qualitative, probably overestimated because of the lower eye sensitivity. Furthermore, the crystals are grown in different flux.

Our experiments show that the gray-tracking peak power threshold strongly decreases when the repetition rate is increased: from 125 MW/cm^2 at 1 kHz to 18 MW/cm^2 at 6.3 kHz, and to a few MW/cm^2 for frequencies greater than 10 kHz. The estimated thresholds are well fitted by an exponential equation: $I_{2\omega}^{\text{threshold}}=A\ \exp(-366.f)$ with f in Hz. The pre-exponential factor A is not yet significant since the thresholds are only qualitative. One can notice that the data

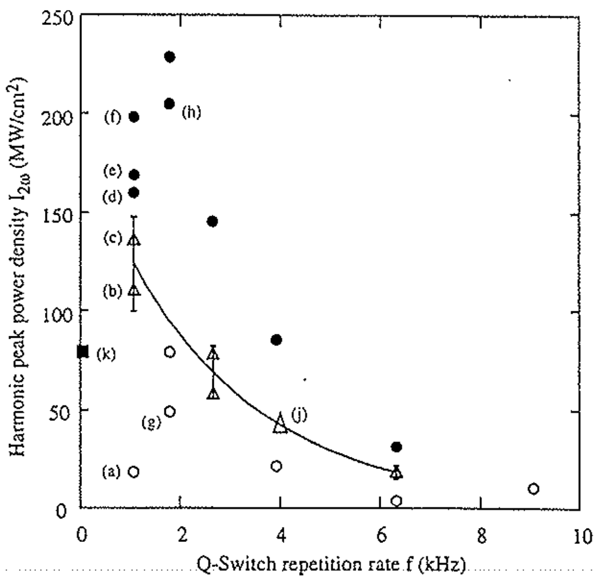

FIG. 1. Gray tracking as a function of the repetition rate, for different harmonic peak power densities during type II 1064 nm SHG in KTP. ○ corresponds to behavior (i), △ to behavior (ii) and ● to behavior (iii). The estimated damage thresholds are symbolized by I. Points (a)–(h) refer to the experiments 1–8 of Table I. (j) is the experiment taken from Ref. 7. (k) is the threshold measured in Ref. 5. The full line is the exponential fit of the present thresholds.

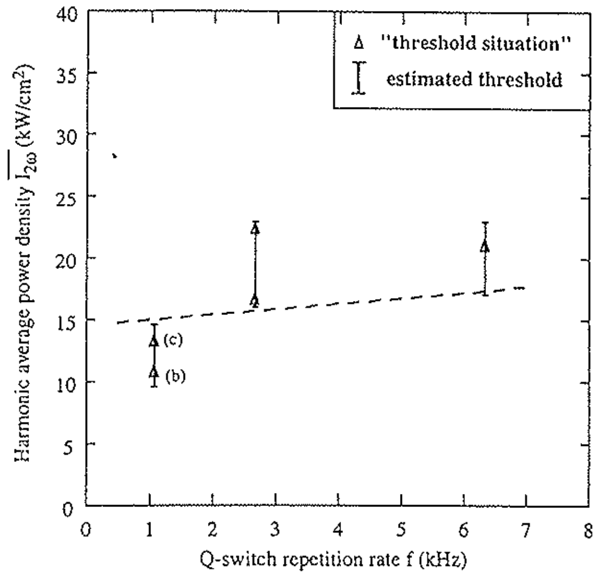

FIG. 2. Average power density at 532 nm corresponding to gray-tracking threshold, as a function of the repetition rate. (b) and (c) refer to experiments 2 and 3 of Table I. The dashed line is a guide for the eyes.

reported from Ref. 7 also fit this equation. The frequency dependence of the threshold is strong. This can be a limitation for high repetition rate applications of KTP.

We show in Fig. 2 the harmonic average power density corresponding to the gray-tracking threshold. It remains quasi constant in the considered frequency range, around 16 kW/cm^2. Among all the measured parameters, it is the only one which does not depend on the frequency in the considered range.

So the average power density is the significant parameter of gray tracking. Furthermore, the observed thresholds correspond to damages which are created after the same illumination duration; this indicates that the harmonic fluence at the threshold does not vary with respect to the frequency in the range 1–10 kHz.

Further experiments will allow us to compare the thresholds of crystals which are grown by the hydrothermal method, and from flux with different chemical compositions. Such results will be interpreted with the impurity analyses and the absorption spectra. The influence of temperature is also fundamental for the comprehension of the gray-tracking mechanism.

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