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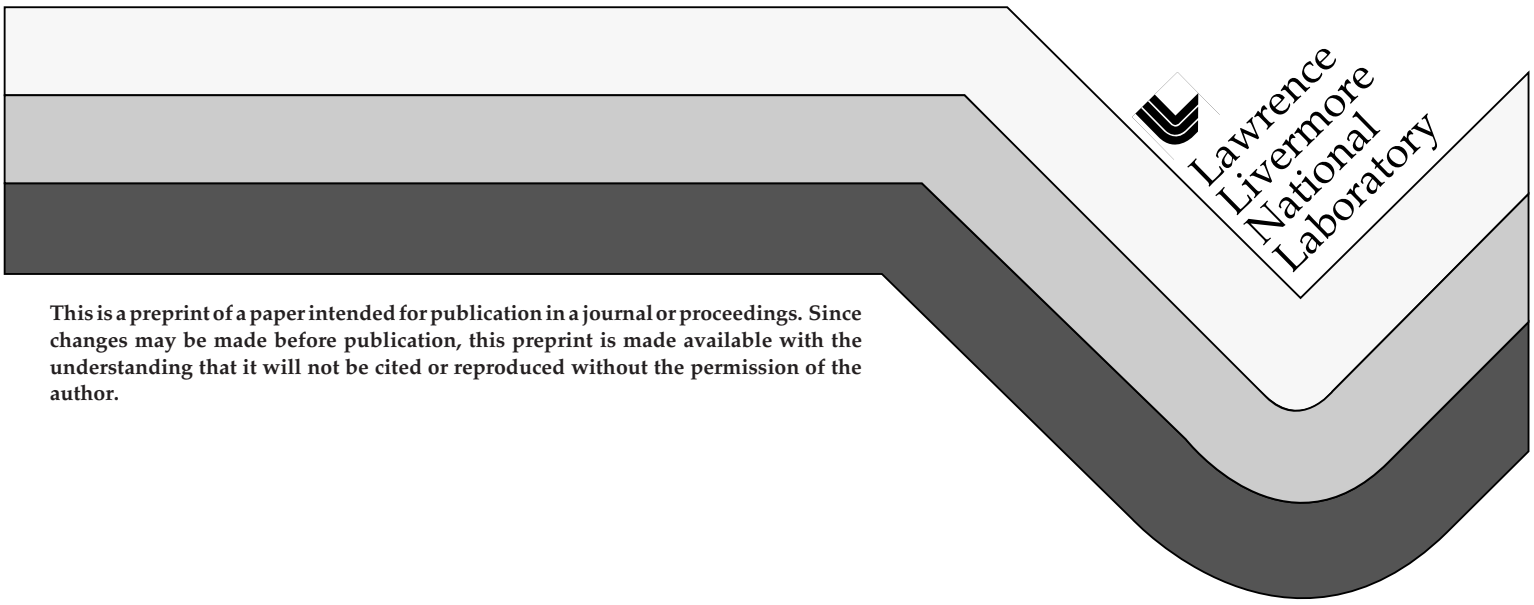

Temperature and spectral investigation of bulk KDP below damage using 355 nm laser irradiation

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ABSTRACT

A spectral and temperature investigation of fast-grown KDP crystals under high fluence, 355 nm laser irradiation is discussed. Pump-and-probe Raman spectroscopy indicate transient changes of the vibrational spectrum. Photothermal deflection experiments provide information on the temporal behavior of the temperature change. The presence of emission in the visible and NIR spectral regions is attributed to the presence of impurities and/or defects in the crystal.

Key words: KDP, Raman scattering, photothermal deflection, damage

1. INTRODUCTION

The continuous development of high power laser systems is accompanied with the need for optical components with enhanced performance characteristics. One of the main problems is to design optical components that have increased damage threshold. In the presence of defects, the physical processes preceding laser induced damage in wide-gap optical materials are even more complex.[1-4] It is currently believed by many researchers that bulk damage in nonabsorbing optical materials is initiated by the generation of free carriers which, in turn, absorb more light and lead to irreversible cascade processes.

In this work, Raman scattering, photothermal deflection and emission spectroscopy techniques are employed to study processes taking place at the sub-damage threshold intensity in fast grown KDP crystals. The experimental results indicate a transient increase of the crystal temperature, emission arising from impurities and/or defects in the crystal and a strong indication of a transient change in the absorption.

2. EXPERIMENTAL SET-UP

The experimental arrangement for the Raman scattering experiments consists of a Q-switched Nd:YAG laser operating at 10 Hz, a CW argon laser, a single grating spectrograph and a liquid nitrogen cooled CCD detector. The 10 ns, 355 nm third harmonic of the YAG laser is focused to ≈1 mm diameter and it used as the pump beam illuminating the sample with ≈5 J/cm^2 along the z-axis of the crystal. The probe laser beam is the 10 ns, 532 nm second harmonic of the YAG laser illuminating the sample with ≈0.5 J/cm^2 with the pump pulses preceding the probe pulse by 10 ns. The probe beam is propagating along the x-axis of the crystal and it is focused to ≈100 μm diameter at the crossing point with the pump beam. The pump and probe beams are overlapping inside the crystal and the Raman scattering signal from the point of overlap of the two beams is spectrally analyzed and recorded using x(zz)y scattering geometry. The samples used were fast-grown KH_2PO_4 crystals.

The laser beam utilized in the photothermal deflection and emission spectroscopy experiments is the third harmonic of a Q-switched, 3 ns pulsewidth, Nd:YAG laser. The arrangement of the photothermal deflection experiments involves overlapping the 355 nm pump beam focused to ≈30 μm diameter with the CW beam of a diode laser focused to ≈85 μm diameter in the bulk of the crystal at an angle of ≈45 degrees. The deflection of the diode laser beam is measured using a quadrant detector and the signal is recorded using a sampling oscilloscope and a box-card integrator. The emission spectroscopy experiments were performed with the sample held at room temperature. The emitted light under 355 nm illumination is spectrally analyzed using a quarter meter spectrograph and recorded using a CCD detector.

3. RAMAN SCATTERING

Fig. 1 shows the Stokes and antiStokes Raman spectra of the 532 nm probe beam when the pump beam is on and when the pump beam is turned off. Three phonon peaks are observed in the x(zz)y geometry arising from the internal modes located at 363 cm^{-1} (v2), at 515 cm^{-1} (v4) and at 915 cm^{-1} (v1).[5-6] The two spectra are overlapping except for the 915 cm^{-1} peak which has higher intensity (the difference in intensity is denoted as ΔI) when the pump beam is

illuminating the sample. The increase of the 915 cm^{-1} peak is observed in both, the Stokes and antiStokes parts of the spectrum. The intensity of the antiStokes 363 cm^{-1} peak remains the same in both cases which indicates that there is no change of the crystal temperature

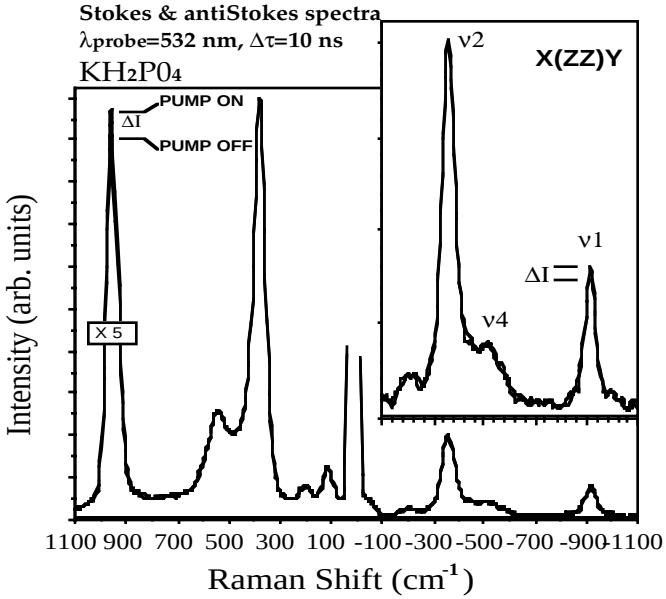

Figure 1. Stokes and antiStokes Raman spectra of the 532 nm probe beam when the pump beam is on and when the pump beam is turned off. The inset shows the antiStokes Raman spectrum in more detail. ΔI denotes the change in intensity of the 915 cm^{-1} mode when the pump beam illuminated the sample.

within the experimental resolution of the Raman scattering system which is limited to approximately a few degrees. The same conclusion is reached when the ratio of the Stokes over the antiStokes intensities of the 915 cm^{-1} modes is used to obtain information regarding the temperature of the sample at the point where the pump and probe beams are overlapping.

Fig. 2 shows the Stokes Raman spectra of the probe beam in more detail when the pump beam is on and when it is turned off. The intensity of the of the 915 cm^{-1} mode is higher when the pump beam is illuminating the sample. The difference in intensity is $\Delta I \approx 0.041\ I_{max}$ where I_{max} is the peak intensity of the 915 cm^{-1} line. The inset shows the digitized intensity difference spectrum after subtraction of the Raman spectrum of the probe when the pump beam is off from the Raman spectrum of the probe when the 355 nm pump beam is present. The peak observed in the difference spectrum is located at 915 cm^{-1}. No additional features are observed in this spectrum.

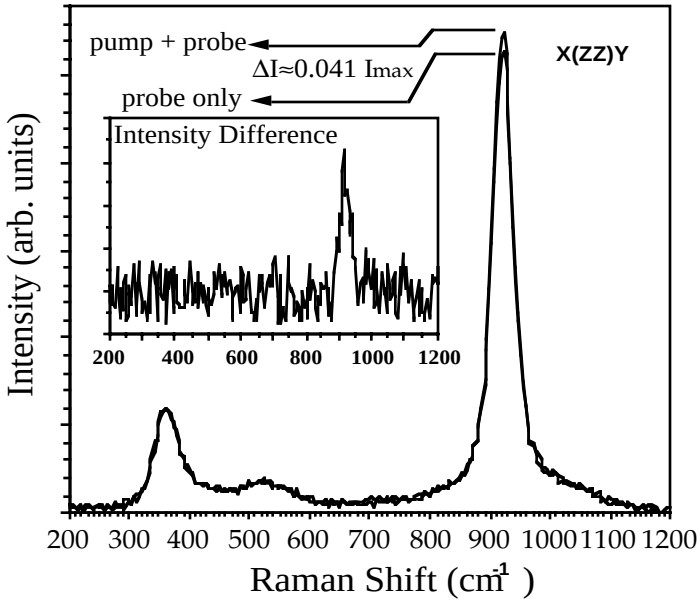

Figure 2. Stokes Raman spectra of the probe beam when the pump beam is on and when it is turned off. The inset shows the difference spectrum after subtraction of the digitized intensity profiles of the two Raman spectra.

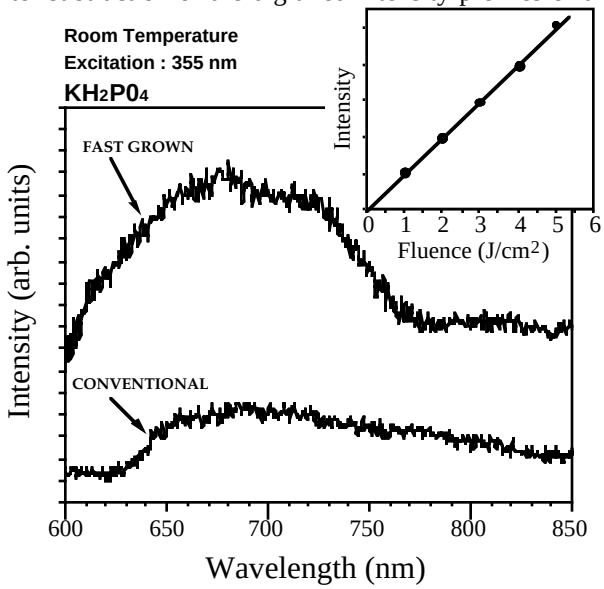

Figure 3. Emission spectra in the 600 nm to 850 nm spectral region from KDP crystals grown using the conventional and fast growth methods under 355 nm illumination. The inset shows the integrated intensity of the emission as a function of the laser power

4. EMISSION UNDER 355 NM ILLUMINATION

KDP crystals grown using the conventional and fast growth methods were illuminated with 355 nm laser light using below damage threshold laser fluences. Fig. 3 shows emission spectra in the 600 nm to 850 nm spectral region. The samples were held at room temperature. The change of the integrated intensity of the emission as a function of the laser power, shown in the inset of Fig. 3, demonstrates a linear dependence, indicating that the emission is due to single photon excitation of impurities in the bulk. The exact information on the composition of these impurities is not yet known. KDP samples obtained from the pyramidal sector exhibit lower emission than from the prismatic sector of the same crystal, indicating a difference in the content of impurity ions in the two sectors. Experiments to determine the damage threshold in the two sectors demonstrated practically no difference in the damage threshold. This suggests that atomic impurity ions in the crystal do not play a significant role in the damage mechanism in KDP.

5. PHOTOTHERMAL DEFLECTION

The photothermal deflection technique has been proven to be a very sensitive method to measure transient variation of local temperature.[7] This technique was employed to study the transient temperature change of the lattice following illumination with high power, 355 nm , laser pulses of 3 ns time duration. Fig. 4a shows the photothermal deflection signal from the pyramidal and the prismatic sectors of the same crystal. The change in intensity may be due in part to the difference in the concentration of impurities in the two sectors and the heat deposited into the local lattice environment following the nonradiative relaxation of the photoexcited impurity ions. The decay time of the photothermal deflection signal reflects the diffusion of the transient localized heat into the rest of the crystal. Fig. 4b shows the dependence of the photothermal deflection signals on the 355 nm power of the laser pulse in a KDP crystal and in a light absorbing neutral density (ND) filter. This dependence is linear for the ND filter due to the single photon absorption processes associated with the heat deposited in the illuminated volume. In the KDP however, the dependence of the photothermal deflection signal on the energy of the laser pulse is not linear. This indicates multiphoton or multistep excitation processes are taking place in KDP in addition to the single photon excitation arising mainly from impurities inside the crystal.

6. DISCUSSION

The increase of the intensity of the 915 cm^{-1} mode when the high power 355 nm illumination beam is present may be attributed to the following possibilities: 1) the presence of stress causing a change of the Raman spectrum, 2) depolarization of the Raman scattering light and spectral component other than the A_1 modes observed and, 3) change of the absorption characteristics following the pump pulse causing resonant Raman enhancement.

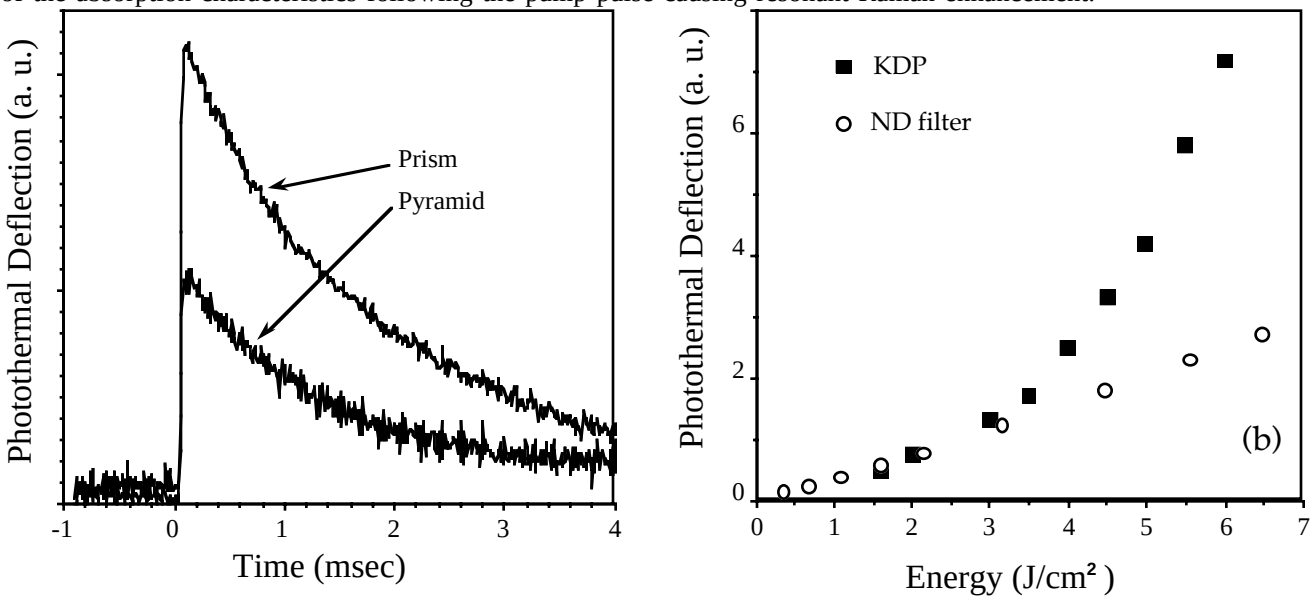

Figure 4. a) Temporal profile of the photothermal deflection signal from the pyramidal and the prismatic sectors of the same KDP crystal. b) The dependence of the photothermal deflection signals to the 355 nm laser power in KDP crystal and in a neutral density filter

It has been shown that when stress is applied in KDP crystals, the Raman scattering spectrum changes and shifting of the position of individual modes is observed.[8] In this experiment, no shifting of the modes is observed within the experimental resolution of the system (≈8 cm^{-1}).

If depolarization is causing the observed changes, one would expect that in addition to the A_1 modes observed in the x(zz)y geometry, the spectral characteristics of the Raman spectrum in the x(zx)y geometry (the main feature of this spectrum is the dominant broad 515 cm^{-1} line) should become more prominent. However, this does not happen. No other alteration than the intensity change of the 915 cm^{-1} line in the x(zz)y Raman spectrum is observed when the 355 nm pulses illuminate the sample. In addition, depolarization will lower the intensity of all A_1 modes rather than increasing the intensity of only one of them as is observed.

A transient change of the absorption might be another reason for the observed changes. Absorption can be due to impurity ions (X) replacing P in tetrahedraly coordinated sites and due to absorption, local modes of the XO_4 could be observed. However, in this case due to the difference in mass between the impurity ion and P, the local modes should be shifted in energy with respect to the PO_4 internal modes.[9] This is not consistent with the experimental observations (see inset of fig. 2). The above discussion indicates that if absorption is the source of the changes in the Raman spectrum, it has to be associated with the PO_4 itself. In this case, based on Albrecht's theory[10], the internal modes of PO_4 that should be most enhanced are the totally symmetric ones and foremost, the tetrahedral 915 cm^{-1} breathing mode which is the only totally symmetric mode in the C_2 site symmetry that originates in a totally symmetric representation in the Td molecular symmetry. This model is consistent with the experimental results, where the 915 cm^{-1} mode is enhanced without any shifting in energy.

The ability of Raman scattering to measure the lattice temperature using the ratio of the intensity of the Stokes over the antiStokes lines of the same mode is limited to few degrees due to insufficient signal to noise ratio. The photothermal deflection technique is much more sensitive in detecting transient temperature variations compared to Raman scattering. The temperature-time profiles measured using the photothermal deflection technique are due to temperature variations smaller than a few degrees. The exact value of the temperature change has not been estimated. We are currently testing a system to perform a three dimensional mapping of the temperature change in KDP crystals based on the photothermal deflection technique to investigate the presence of "hot spots" inside the crystal that may be responsible for lowering the laser damage threshold.

7. CONCLUSION

In conclusion, the Raman scattering, photothermal deflection and emission spectroscopy investigation of fast-grown KDP crystals under high fluence, 355 nm laser irradiation allowed for the investigation of physical processes taking place at below damage threshold. The Raman scattering experiment indicates that the change of the local temperature is less than few degrees while it is strongly suggestive of a transient change in the absorption. Using the photothermal deflection technique, the dynamics of the transient temperature variations indicates that in KDP the temperature increase is in part due to multistep excitation. Emission spectroscopy measurements have revealed the presence of impurities. The degree that these processes are related to the damage mechanism in KDP is subject to further investigation.

8. ACKNOWLEDGMENTS

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