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Induced Gain in Cr,Nd:GSGG by Pulsed X-Ray Radiation

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Induced gain in Cr,Nd:GSGG by pulsed X-ray radiation

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Abstract

A normalized X-ray induced gain coefficient, $1.48 \times 10^{-3} \text{ cm}^{-1}/\text{krad}$, has been determined for Cr,Nd:GSGG. Hermes III, a 20 ns, 2 MeV X-ray source, is used to irradiate the sample. Doping levels of 1×10^{20} and $2 \times 10^{20} / \text{cm}^3$ respectively, for Cr and Nd are used. A proposed heuristic model in which excited Gd transfers its excitation to Cr which then transfers its excitation to Nd is described.

Acknowledgments

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Introduction

Recently it has been shown that Cr,Nd:GSGG displays gain at 1061 nm when it is irradiated with a 20 ns pulse of 2 MeV X-rays.^{1,2,3} It has been noted that the peak gain occurs 10-20 μ s after the X-ray pulse. This delay has been associated with the excitation transfer time from Cr³⁺ to Nd³⁺. Gain has been demonstrated by both a 1061 nm probe beam and by lasing action from a laser cavity. The estimated output energy from the laser experiment is about 200 mJ when the laser rod is exposed to a 70 krad dose of X-rays. Normalized gain coefficients have been obtained from both the laser experiment and the probe experiments. A corner cube has been used in the laser experiment and the normalized gain coefficient determined by equating the gain to the losses in the cavity while operating near threshold. The primary uncertainties in this gain estimate are the loss in the corner cube and the exact threshold. A normalized gain coefficient of 0.00088 cm⁻¹ krad⁻¹ has been determined in this case.³ In the probe beam experiments, the sample is from the end of the boule where the doping levels are uncertain. A normalized gain coefficient of 0.0067 cm⁻¹ krad⁻¹ has been determined for this case. Contributions to the signal by spontaneous emission have been neglected here and consequently, the estimate of the gain coefficient is expected to be too high. The uncertainty in the doping levels and the omission of the spontaneous emission corrections leave the exact normalized gain coefficient in doubt for these earlier experiments. This study is a probe beam experiment in which corrections for spontaneous emission have been made, and hopefully accurate normalized gain coefficient derived. Also, one sample is taken from laser rod material whose doping levels has been measured accurately.

The host laser material, gadolinium scandium gallium garnet (Gd₃Sc₂Ga₃O₁₂), is a member of the garnet family. It commonly referred to as GSGG. The molecular formula for this general class of materials is C₃A₂D₃O₁₂.⁴ The C ion is located in a dodecahedrally coordinated site, the A ion in an octahedrally coordinated site, and the D ion in a tetrahedrally coordinated site. When this material is doped with Nd, the Nd can substitute for the C ion. In this location, the interaction between the Nd ion and the host material is small so that the atomic lines of the Nd ion are relatively sharp. Doping this material with Cr can result in the Cr substituting for the A ion. In this case, the Cr ion experiences an electric field with octahedral symmetry and the energy levels are labeled accordingly. A measure of the uniformity of the dopants is given by their segregation coefficients.⁴ The segregation coefficients for Cr and Nd are respectively, 0.96 and 0.65. Figure 1 is a schematic of the energy levels of the Cr and Nd ions in the host material..

Luminescence and stimulated emission studies of Nd-doped GSGG were first reported in 1975.⁵ Shortly thereafter several groups reported laser studies of Nd:GSGG sensitized with Cr.^{6,7,8,9} The sensitized material, Cr,Nd:GSGG, has approximately three times the slope efficiency as unsensitized Nd:YAG when flashlamp pumping is used.¹⁰ This increase in efficiency is attributed to the broad absorption bands of the Cr ion in the visible part of the spectrum which matches the output of the flashlamp. Also, there is efficient transfer of the excitation energy from the Cr³⁺ ion to the Nd³⁺ ion.^{6,8} The transfer

of excitation from a Cr^{3+} ion in the $^4\text{T}_2$ and ^2E states to a Nd^{3+} ion is indicated by the broad arrow in Figure 1. Efficient mixing occurs between these two states through spin orbit coupling because of the small energy gap between them. This transfer process is less efficient in Cr, Nd:YAG, and there is very little gain in the laser output when Nd:YAG is sensitized with Cr.¹⁰

The long-term radiation hardness of Cr,Nd:GSGG lasers is well documented in the literature. Doses of 10 Mrad of ^{60}Co γ -rays do not affect the laser's performance.¹¹ A 1 Grad dose of γ -rays decreased the output by about 5%.⁴ This is in contrast to a Nd:YAG laser whose output is dropped by an order of magnitude by a 1 Mrad dose of γ -rays.¹¹ It has been shown that solarization by γ -ray and UV radiation can be avoided by co-doping GSGG with Cr and Nd.¹¹ Similar studies using electron and proton irradiation have proven Cr,Nd:GSGG is a radiation hard material.⁴ These characteristics are only observed when both Cr and Nd are present.¹² It has been shown that this material has a high optical damage threshold even after a 100 Mrad γ -ray doses.¹³ All of these studies are for long term radiation effects and do not address the transient radiation properties for the time frame 0.1 to 1000 μs after a short radiation pulse. In the experiments of the present report, this is the time frame studied.

Experimental methods

The general optical setup has been described before¹⁴ and will not be repeated in detail here. Two laser beams are created by splitting a single beam; they are then transmitted through separate optical paths. Each of these probe beams is imaged on the end of a fiber optic cable and is transmitted through the fiber to a high intensity X-ray chamber. At the end of these fibers are collimator lenses which collimate the beams. Each beam is propagated through the air for about 1 m, through a sample that is being probed, and then returned through the sample by a retro-reflecting mirror. It is then re-imaged onto the end of the fiber optic cable by the collimator lens. Both beams return to the shielded instrument room, where the laser is located, by the fiber optic cables described above. A beam splitter picks off about 50% of a return beam and passes it to the detector. With this arrangement two samples can be probed simultaneously.

Two samples of Cr,Nd:GSGG were used for the gain measurements. A large bulk sample described previously^{1,2,3} has Cr and Nd densities of 1×10^{20} and $2 \times 10^{20} / \text{cm}^3$, respectively. Because this sample has been taken from the end of a boule, it is suspected that the densities could be in error. This sample is $25.4 \times 25.4 \times 12.7$ mm and the large faces are polished to an optical finish. The probe beam is incident on the large faces so that it passes through the sample along the 12.7 mm dimension. No AR coating is applied to the faces of this sample. The second sample has been taken from a laser rod and the Cr and Nd densities have been accurately determined to be the values given above. It is 6 mm in diameter and 6 mm long with both end faces of the rod polished to an optical finish. The end faces are AR coated to reduce the Fresnel reflections. This second sample is mounted in $25.4 \times 25.4 \times 12.7$ mm Teflon block. Both samples are placed on pedestals near the retro-reflectors described earlier.¹⁴

Results and conclusions

Figure 2 is the transmittance of the laser rod when subjected to a 68.9 krad (CaF_2)* dose of 2 MeV X-rays. The 5 - 6 % decrease in transmittance at the later times is due to induced absorption in the combination of the collimator and the fiber optic cable. This absorption must be taken into account when determining the gain. An independent experiment without the laser rod revealed that the recovery time of the combination is much longer than the time frame shown in Figure 2, and that the induced absorption can be considered constant for the time frame shown. It can be seen that the peak of the transmittance occurs 10-20 μs after the radiation pulse, which is consistent with the transfer time of excitation from Cr to Nd.⁶

The signal measured in Figure 2 has two components, one due to spontaneous emission and the other due to gain of the probe beam. The part due to spontaneous emission has been determined by turning the probe laser off and subjecting the laser rod to a X-ray radiation pulse. The peak of the spontaneous emission appropriately scaled for dose is subtracted from the peak of the total signal and the new transmittance determined at the peak. The small signal gain can be determined from this peak transmittance and the double pass length of the sample. Similar measurements have been made on the 25.4 $\times$ 25.4 mm sample. Normalized small signal gains of 1.67×10^{-3} and $2.06 \times 10^{-3} \text{ cm}^{-1}/\text{krad}$ (CaF_2)* have been found for the laser rod and the bulk GSGG sample, respectively. The difference is within the uncertainties in the concentrations of the Nd and Cr in the GSGG as determined by the segregation coefficients of Cr and Nd.⁴ At this time it is not known which of these two elements is causing the difference.

It has been pointed out earlier that the mechanism for the excitation of the Nd^{3+} ion in GSGG is not understood, but a simple heuristic model will be given here. A detailed model for the excitation mechanism is beyond the scope of this study. Further experiments are necessary to validate the heuristic model given below. Experiments in which the excited levels of the Gd^{3+} ions in the ^6P states are probed should be performed to determine the accuracy of the model.

When the 2 MeV photons are incident on the GSGG, the primary interaction is the generation of Compton electrons. These Compton electrons then generate secondary electrons throughout the crystal, which then excite the individual ions in the crystal. Since Gd has an atomic number of 64, Ga an atomic number of 31, Sc an atomic number of 21, O an atomic number of 8, and the molecular formula for GSGG is $\text{Gd}_3\text{Sc}_2\text{Ga}_3\text{O}_{12}$, it is intuitive that Gd with the most electrons has the highest probability of being excited by the X-ray irradiation. Certainly for the original 2 MeV X-rays this is true, since the binding energies of the electrons are small compared to the X-ray energy, and only the

* The dose indicated here is for CaF_2 ; a correction factor of about 1.13 is required to determine the true absorbed dose. The normalized gains have to be reduced accordingly.

number of electrons is important. This is also true of the high energy secondary electrons. The picture becomes less clear when the energies of the secondary electrons approach 0.1 MeV, which is the approximate magnitude of the binding energies of the inner electrons of the atoms. Recombinations and cascading effects cloud the picture even further so that it is difficult to predict which energy levels in the crystal may be excited. However, it seems reasonable that the Gd^{3+} ion would be more likely to be excited than the other ions in GSGG, and this could be an important step in the excitation mechanism. Assuming this argument has merit, and using the reported result that a Cr dopant in GSGG can be excited by Gd^{3+} ions in the 6P manifold of energy levels,¹⁵ it is plausible that Cr^{3+} can end up in an excited state. It has been shown that excited Cr^{3+} transfers its excitation to Nd^{3+} in Cr, Nd:GSGG. An excited Gd^{3+} ion in the 6P manifold transfers its excitation to a Cr^{3+} ion in about 300 ns which is small compared to the 10 μs transfer time from Cr^{3+} to Nd^{3+} .⁶

In conclusion, a normalized X-ray induced gain coefficient of $1.48 \pm 0.15 \times 10^{-3}$ $cm^{-1}/krad$ (GSGG) has been determined for Cr and Nd doping levels of 1×10^{20} and 2×10^{20} , respectively. A heuristic model based on the transfer of excitation from Gd^{3+} to Cr^{3+} and then to Nd^{3+} is proposed as important steps in the excitation mechanism. Other cascading steps in the upper excited levels of Gd^{3+} are unknown.

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Figure 1, Energy levels of Cr^{3+} and Nd^{3+} in GSGG

Figure 2: Uncorrected transmittance of small laser rod. Corrections for spontaneous emission and induced absorption in the combination of the fiber optic cable and collimator must be made to get true transmittance

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