High-gain nondegenerate two-wave mixing in Cr:$YAlO_3$

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A gain of 22 times was obtained by nondegenerate two-wave mixing in Cr:$YAlO_3$. To our knowledge, this is the largest cw two-wave mixing gain obtained in a bulk solid-state nonphotorefractive material. The measured gain appears to be limited by beam breakup that is due to spatial nonuniformities in the nonlinear refractive-index change that are the result of inhomogeneities in the crystal. Predictions based on our data indicate that gain in excess of 200 times should be possible in Cr:$YAlO_3$ if a homogeneous crystal can be obtained.

Recently we demonstrated a gain of six times by nondegenerate two-wave mixing (NDTWM) in chromium-doped yttrium aluminate (Cr:$YAlO_3$).[1] Here we report new experiments in which we achieved a gain of 22 times by increasing both the interaction length and the doping. To our knowledge, this is the largest cw NDTWM gain obtained in a solid-state nonphotorefractive material. The large gains possible in this material should permit the development of cw oscillators and self-pumped conjugators with properties different from those employing photorefractives.

Experiments demonstrating high-gain NDTWM in new solids by use of low-power cw lasers constitute an essential step in establishing the practicality of alternative materials for applications of nonlinear optics in areas such as signal processing. Availability of high gain can bring about rapid progress in experimental research, as occurred with the discovery of photorefractives such as barium titanate ($BaTiO_3$). To our knowledge, however, no one has demonstrated gain comparable with that of photorefractives by using a solid-state bulk (as opposed to guided-wave) Kerr medium with a cw source. We report properties of a candidate material, Cr:$YAlO_3$, that may significantly advance experimental efforts on nonlinear optics in Kerr-like media, much like the way in which basic studies of $BaTiO_3$ initiated widespread study of nonlinear optics in photorefractives. Motivation for this study was provided by the expectation of new properties that should result from the different nonlinear mechanism (Kerr-like versus photorefractive), such as improved dynamic range in mutually pumped phase conjugation,[2] that is important in wave-front-matched heterodyne receivers.[3]

In some metal-ion-doped insulators, such as ruby, metastable states result in low saturation intensities and correspondingly large optical nonlinearities. The origin of the nonlinearity was previously determined to be the result of light-induced population changes and the difference in the susceptibility between the ground and metastable states.[4] Interest in these materials stems from the fact that they are convenient solid-state materials with large optical nonlinearities that can be utilized with relatively low-power cw lasers. Photorefractives also meet these conditions and have demonstrated two-wave mixing gain of $>10^3$, but the differences in the nonlinear mechanisms make investigation of the metal-ion-doped insulators of interest because of their potential advantages. As one example, the beam ratio for which mutually pumped phase conjugation will take place in photorefractives is limited, but this is not the case in Kerr materials.[2] Our search of the literature addressing nonlinearities in metal-ion-doped insulators indicated that Cr:$YAlO_3$ stands out as the material having the largest potential index change for a given amount of absorption, i.e., the largest value of $n_2 I_S/\alpha$, where I_S is the saturation intensity, n_2 is the optical Kerr coefficient, and α is the absorption coefficient.[5] We therefore chose this material for experimental attempts to obtain high two-wave mixing gain.

As grown, Cr:$YAlO_3$ has a color-center absorption that competes with the absorption responsible for the optical nonlinearity. To remove the color centers, we heated as-grown material in a forming gas atmosphere.[6] Figure 1 shows absorption spectra for the 12-mm-thick end of a boule of Cr:$YAlO_3$ as grown (dotted curve), after a first anneal (dashed curve) in 95% N_2/5% H_2 at 1050 °C for 8 h with a flow rate of 250 cm^3/min and a ramp rate of 1 °C/min, and after a second anneal (solid curve) at 1300 °C for 24 h with the same atmosphere and the same flow and ramp rates as used in the first anneal. After the

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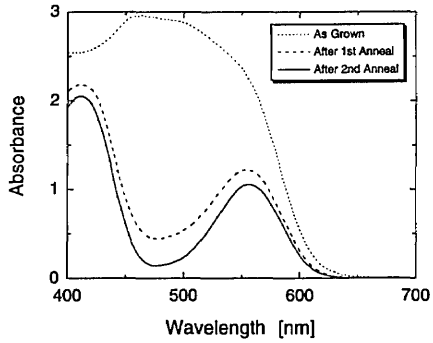


Fig. 1. Cr:$YAlO_3$ absorption spectra showing the bleaching of color centers by heat treatment.

second anneal the boule was fabricated into several crystallographic-cut cubes, one of which was used in these experiments. Microprobe measurements indicated that the Cr concentration was 0.12 wt.%.

Figure 2 is a simplified schematic of the experiment used to measure NDTWM gain.[7] Light from an Ar laser at 514.5 nm is split by a beam splitter into a weak probe beam $I_2(0)$ and a strong pump beam $I_1(0)$. We shift the frequency of the pump with respect to the probe by reflecting it from a mirror mounted to a piezoelectric transducer and driven by a triangle-wave voltage source. This results in a moving interference pattern in the crystal and a population grating that lags in phase because of the finite response time. As in photorefractives, this phase shift is responsible for the energy exchange. The sign of the phase shift depends on the sign of the frequency shift and determines the direction of energy flow. Amplification of the probe results when the mirror moves so that the pump is upshifted, and attenuation results when the mirror moves in the opposite direction so that the pump is downshifted. The triangle-wave frequency and thus the pump frequency was varied to maximize gain. This maximum occurs for a frequency shift of ~5 Hz, consistent with the theoretical behavior described by[7,8] $1/(2\pi\tau) = 4.8$ Hz, where τ is the metastable-state lifetime of 33 ms. Steady state is achieved since the period of our triangle wave (~7 s) greatly exceeds τ.

To obtain the intensities required for high gain, we focus the beams to a $1/e^2$ waist of 210-μm radius at the crystal. We ensure a high degree of overlap in the crystal length $L = 9.1$ mm by having the beams intersect at the small angle of 0.02 rad. The beams were incident nearly parallel to the ⟨001⟩ direction and s polarized perpendicular to the ⟨$\bar{1}$10⟩ direction. This polarization and crystal orientation yield the highest peak in the absorption spectrum at 560 nm and the lowest minimum in the residual absorption between peaks at 475 nm. Thus we believe that this configuration yields the greatest interaction with the absorption responsible for the optical nonlinearity and the least interaction with residual color centers. Depletion of the pump is avoided by use of a large input beam ratio, i.e., $I_1(0)/I_2(0) = 1000$.

Figure 3 shows the gain $I_2(L)/I_2(0)$ as a function of the pump intensity. The solid curve is a fit of

$$\frac{I_2(L)}{I_2(0)} = \exp(-\alpha L) \times \exp\left(\left[\frac{1-\exp(-\alpha L)}{\alpha L}\right]\left\{\frac{2\pi n_2 I_1(0)L/\lambda}{[1+I_1(0)/I_S]^2}\right\}\right), \quad (1)$$

with $\alpha L = 0.69$, $\lambda = 514.5$ nm, $I_S = \hbar\nu/\sigma\tau = 1.27$ kW/cm^2, where $\hbar$ is Planck's constant, ν is the optical frequency, σ is the absorption cross section, and τ is the metastable-state lifetime. With n_2 as the only adjustable parameter, the fit yields $n_2 = 3.2 \times 10^{-7}$ cm^2/W. The nonlinearity that is due to population effects dominates in both beam coupling and self-focusing because the n_2's for the corresponding thermal effects are at least two orders of magnitude lower. We can derive Eq. (1) from Ref. 8 by including the effect of saturation, which reduces the nonlinear index by a factor $[1 + I_1(0)/I_S]^{-2}$, for $I_1(0) \gg I_2(0)$.

The inset in Fig. 3 shows a plot of the gain for the range from zero to 400 W/cm^2. With the previously stated parameters and n_2 from the fit for the lower-intensity range, the curve in the inset represents a theoretical projection based on Eq. (1). A gain of 22 times was obtained at approximately 400 W/cm^2, but the measured gain does not increase with intensity as much as theory predicts. (Theory predicts a gain of 200 at 400 W/cm^2.) We believe this is due to beam breakup resulting from spatial nonuniformities of the nonlinear refractive-index change in the interaction region. As evidence, Fig. 4 shows the transmitted pump *with the probe off* (a) at low intensity and normal exposure (0.6 W/cm^2, 1/100 s) indicating the transmitted beam size, (b) at low intensity and at a longer exposure (1.4 W/cm^2, 1 s)

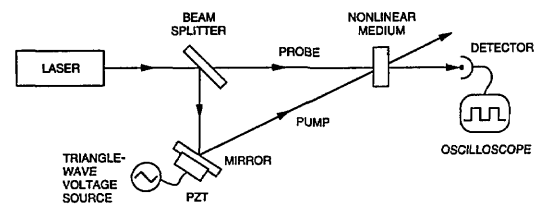


Fig. 2. Simplified schematic of the experiment used to measure NDTWM. PZT, piezoelectric transducer.

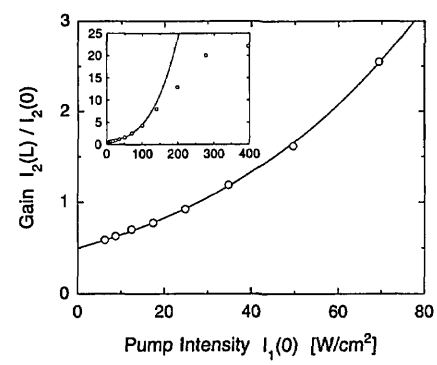
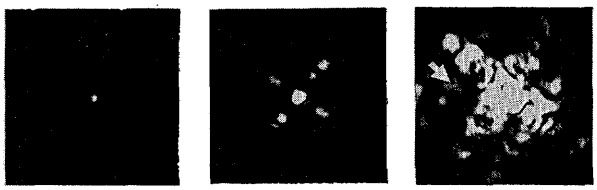


Fig. 3. Gain versus pump intensity. The solid curve is a fit of Eq. (1). The inset shows that, at high intensity, the measured gain does not increase with intensity as much as theory predicts. We believe this is due to beam breakup resulting from spatial nonuniformities of the nonlinear refractive-index change.



(a) (b) (c)

Fig. 4. Transmitted pump with the probe off (a) at low intensity and normal exposure indicating the transmitted beam size, (b) at low intensity and long exposure showing an X-shaped pattern of linear scattering from crystal striae, and (c) at high intensity and an exposure equivalent to that in (b), showing the beam breakup. For all photographs the plane of incidence is the horizontal.

to reveal the X-shaped pattern of weak linear scattering that is believed to be the result of scattering by crystal striae, and (c) at high intensity and an exposure equivalent to that in (b) (140 W/cm^2, 1/100 s) showing the nonlinear scattering that is due to nonuniformities in the nonlinear refractive-index change.

Spatial nonuniformities of the nonlinear refractive-index change in the interaction region can be the result of shadows or distortions from surface or bulk defects or from spatial variations in the Cr-ion concentration. Photographs of the crystal illuminated by a low-intensity collimated beam show radial and concentric patterns thought to be Cr striae. Images of a low-intensity pump beam inside the crystal also indicate that the striae have a significant effect on the beam profile. Beam breakup can result both from variations in Cr concentration, yielding a variation in n_2, and from intensity variations, yielding variations in the induced nonlinear index change.

Qualitative behavior indicated that the scattering at high intensity was nonlinear, but to confirm this we measured the intensity of a small region of the scattering as a function of the pump intensity. An aperture was closed down to a diameter such that, at low intensity, it would pass one half of the pump that transmitted through the crystal, and then it was placed in the speckle of the scatter indicated by the arrow in Fig. 4(c). Measurements indicated that the nonlinear portion of the scatter had a cubic dependence on the pump intensity.

We made the following observations concerning the scattering: (1) the strength of the nonlinear scattering depended on feedback into the crystal and (2) a threshold existed above which the speckle pattern of the scattering was dynamic. An experiment examining the interference between a portion of the transmitted pump and a speckle did not detect any frequency shift. This indicated that the dynamic scattering at high intensities was not due to beating between frequency-shifted stimulated scattering resulting from NDTWM gain and some linear scattering from crystal inhomogeneities. When the crystal was oriented to retroreflect the pump or when an external mirror was used, the strength of the nonlinear scattering increased and the threshold of dynamic scattering decreased. To obtain the highest gains in the NDTWM measurements we rotated the crystal away from normal to reduce the nonlinear scattering. An angle of incidence for the pump of 0.03 rad yielded the largest gains. Further reductions in the feedback resulting from the rear surface reflection might improve the gain.

In conclusion, we demonstrated a gain of 22 times by NDTWM in Cr:$YAlO_3$. The gain appears to be limited by beam breakup resulting from spatial nonuniformities in the nonlinear refractive-index change that are the result of inhomogeneities in the crystal. Theory predicts that gain in excess of 200 times should be possible in a homogeneous crystal. Thus, like photorefractives, Cr:$YAlO_3$ is a convenient solid having the potential for high gain by use of cw lasers. Interest in Cr:$YAlO_3$ stems from the difference in nonlinearity (Kerr-like versus photorefractive) and the resulting potential for new properties rather than from any improvement in sensitivity or speed. Indeed, the sensitivity of Cr:$YAlO_3$ ($\sim 10^{-11}$ m^3/J) is 2 orders of magnitude below that of $BaTiO_3$. Nor is Cr:$YAlO_3$ particularly fast; the grating decay time in Cr:$YAlO_3$ is determined by the metastable lifetime of 33 ms, and this is comparable with the fastest grating decay times obtained in $BaTiO_3$ under moderate illumination. Although speed increases with intensity in photorefractives, it is not clear that they can withstand the intensities used with Cr:$YAlO_3$, making a direct comparison difficult. Gain is determined by the index change, and the maximum possible index change in Cr:$YAlO_3$ ($\Delta n_{max} \sim n_2 I_S = 4 \times 10^{-4}$) is comparable with that of $BaTiO_3$. In addition to its having potential for new properties resulting from the different nonlinear mechanism, we note that Cr:$YAlO_3$ is only one example of a large class of metal-ion-doped insulators. Admittedly, it has the best performance to date, but an effort to develop metal-ion-doped insulators as nonlinear materials that is as extensive as that undertaken for photorefractives may yield new materials with improved figures of merit.

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