Simultaneous, multiple wavelength lasing of (Ho, Nd):$Y_3Al_5O_{12}$

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(Received 15 June 1987; accepted for publication 25 August 1987)

Simultaneous lasing of both Ho^{3+} and Nd^{3+} in the same crystal of yttrium aluminum garnet (YAG) is reported. The crystal was doped with 10% Ho^{3+} and 1% Nd^{3+} ions. Lasing occurred at 2.940 and 3.011 μm due to Ho^{3+} ion transitions and at 1.064 μm due to a Nd^{3+} transition. Appropriate mirrors produced simultaneous lasing at 1.064 and 1.339 μm due to Nd^{3+} ion transitions. The fluorescent lifetimes of both the Nd^{3+} $^4F_{3/2}$ and Ho^{3+} $^5I_7$ states were significantly lower in the doubly doped material than in Nd:YAG and Ho:YAG. This indicates very strong ion-ion interactions in the (Ho, Nd):YAG crystal.

Holmium has been reported to lase in yttrium aluminum garnet (YAG) in the 3-μm region.[1,2] This lasing is due to a transition from the Ho^{3+} $^5I_6$ to the $^5I_7$ level. However, the $^5I_7$ state has a much longer fluorescence lifetime than the $^5I_6$, and the lasing transition is self-terminated. Soviet researchers have observed this behavior in high-concentration holmium garnets and in yttrium orthoaluminate.[3] The (3.011 ± 0.003) μm lasing observed in our laboratory from 15% Ho:YAG clearly shows self-termination. This effect limits the duration of the laser pulse and the efficiency of the lasing process. Kaminskii *et al.* suggested that Nd^{3+} could be used to cross relax the Ho^{3+} $^5I_7$ level and thereby reduce or eliminate the problem of self-termination.[1] We have pursued this idea, and report, for the first time to our knowledge, on the multiwavelength lasing properties of a YAG crystal doped with 10% Ho^{3+} and 1% Nd^{3+} ions. Simultaneous laser action was observed at 2.940 and 3.011 μm by the Ho^{3+} ions, and at 1.064 μm by the Nd^{3+} ions. While the 3.011-μm wavelength has been reported in Ho:$YAlO_3$, we believe this is the first report of the 3.011-μm laser emission in YAG. By changing to mirrors which had nearly equal reflectance at 1.34 and 1.06 μm, simultaneous lasing was observed on both of these transitions, contrary to typical Nd:YAG laser properties.

The rationale behind the suggestion made by Kaminskii is seen by an examination of the energy levels of the Ho^{3+} and Nd^{3+} ions in YAG, shown in Fig. 1.[4] The laser transitions relevant to this work are indicated by the arrows. The Nd^{3+} $^4I_{13/2}$ level lies approximately 750 cm^{-1} below the Ho^{3+} $^5I_7$ state. This may allow energy transfer from the holmium to the neodymium ions, reducing the bottleneck which normally occurs as a result of the long $^5I_7$ lifetime. Measurements of the various state lifetimes involved in the Ho^{3+} laser transitions were made in 15% Ho:YAG and in (10% Ho, 1% Nd):YAG. The lifetime of the Nd^{3+} $^4F_{3/2}$ was determined in 1% Nd:YAG and in (10% Ho, 1% Nd):YAG. These data are listed in Table I. The drastically lower lifetimes of the Ho^{3+} $^5I_7$ and Nd^{3+} $^4F_{3/2}$ levels in the doubly doped material indicate that very strong interactions occur between the holmium and neodymium ions in this crystal.

The entries in Table I were obtained by exciting fluorescence in the crystals with a frequency-doubled, *Q*-switched Nd:YAG laser operating at 532 nm. Wavelength selection was accomplished using a 0.275-m monochromator, and the fluorescence decay was detected with a cooled InSb detector whose response time was < 1 μs. The monochromator was calibrated with the 1.064-μm emission from a Nd:YAG laser, and the instrumental bandwidth was found to be ± 0.003 μm. A digital processing oscilloscope recorded the fluorescence signals, which were then transferred to an HP9825A computer for storage and processing.

The lifetimes were derived from data such as those shown in Fig. 2, where the log of the fluorescence signal is plotted versus time. Up to 400 decay signals were averaged to obtain an improved signal-to-noise ratio. As Fig. 2 shows,

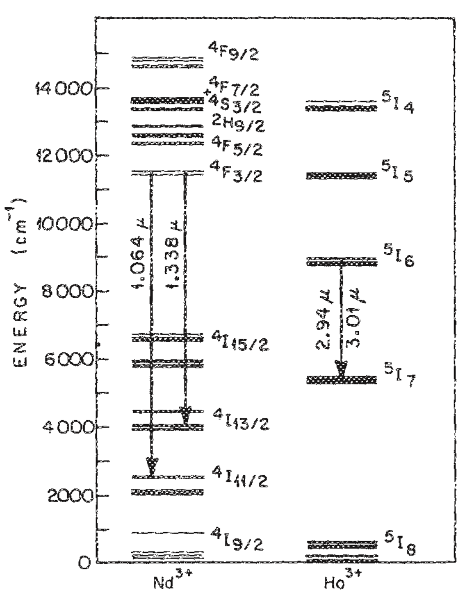

FIG. 1. Energy level diagram for Ho^{3+} and Nd^{3+} in YAG. Laser transitions are indicated by the arrows.

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TABLE I. Lifetimes of the states of Ho^{3+} and Nd^{3+} in (Ho, Nd):YAG and Ho^{3+} in Ho:YAG and Nd^{3+} in Nd:YAG.

State	Material	Lifetime (μs)
$^5I_6$ of Ho^{3+}	15% Ho:YAG	47 ± 3
$^5I_6$ of Ho^{3+}	(10% Ho, 1% Nd):YAG	41 ± 3
$^5I_7$ of Ho^{3+}	15% Ho:YAG	5500 ± 500[a]
$^5I_7$ of Ho^{3+}	(10% Ho, 1% Nd):YAG	170 ± 10
$^4F_{3/2}$ of Nd^{3+}	1% Nd:YAG	237 ± 10
$^4F_{3/2}$ of Nd^{3+}	(10% Ho, 1% Nd):YAG	8.5 ± 0.8[a]

[a] These entries are the initial $1/e$ decay time of a process which does not appear to have a single exponential decay rate.

the Nd^{3+} $^4F_{3/2}$ decay in (Ho, Nd):YAG is nonexponential, exhibiting a continuous curvature. This continuous change in the decay rate of the fluorescence was followed out to beyond 200 μs by using appropriate delays on the oscilloscope. The value given for the decay time in Table I is the initial $1/e$ decay time. This behavior was observed from 1.064 and 1.339 μm emission, which both originate from the $^4F_{3/2}$ level. The decrease in the Nd^{3+} lifetime is evidence of significant energy transfer from Nd^{3+} to Ho^{3+} in this crystal.

The Ho^{3+} $^5I_6$ lifetime has been reduced by only 15% due to the presence of the Nd^{3+}, while the $^5I_7$ lifetime was reduced by a factor of 30 or more, as shown in Table I. The decay of Ho^{3+} $^5I_7$ in 15% Ho:YAG is nonexponential. It was not possible to precisely measure the Ho^{3+} $^5I_5$ lifetime due to detector sensitivity and response time. However, the data that were obtained showed that the lifetime of this level was < 5 μs. The decay processes are being studied further.

All lasing tests were conducted in a silver-coated double-elliptical pump cavity with xenon flashlamps. The arc length was 100 mm with a pump pulse duration of 175 μs full width at half-maximum. Due to the short upper level lifetimes of both Nd^{3+} and Ho^{3+} in (10% Ho, 1% Nd):YAG the long pump pulse may have resulted in inefficient pumping. The laser material was grown using the Czochralski technique. A rod 6.35 mm in diameter and 70 mm in length was used, although the pumped rod length was 64 mm. The input energies in Fig. 3 represent energy input to the lamps, corrected for the mismatch between arc and pumped rod lengths. The rod was prepared with flat, parallel, uncoated faces. Several mirror sets were used for the lasing tests, but none was specifically designed for multiwavelength operation. All mirrors used were flat.

When a mirror designed for 90% reflection at 2.94 μm was used in conjunction with an enhanced silver total reflector designed for 100% reflection at 2.94 μm, simultaneous lasing was observed at 2.940, 3.011, and 1.064 μm (all are ± 0.003 μm), with the output energies indicated in Fig. 3. A temporal shift of the lasing from 2.940 to 3.011 μm was observed. This is similar to that reported by Soviet researchers for Ho^{3+} and Er^{3+} in other hosts.[3,5] The 2.940-μm lasing always precedes the 3.011-μm transition in time. Due to the longer lifetime of the lower state, the lower Stark levels fill during the initial lasing process and the gain decreases. Lasing then switches to the longer wavelength transition. Ho^{3+} lasing at 3.011 μm has not previously been reported in YAG.

For comparison, in 15% Ho:YAG, only the 3.011-μm emission was obtained, and all lasing ceased prior to the peak of the pump pulse. This is in contrast to the (Ho, Nd):YAG, where 3.011 μm emission switched on near the end of the 2.940-μm lasing, and continued past the peak of the pump. A combined total output energy of 41 mJ was obtained from the (10% Ho, 1% Nd):YAG material at 2.940 and 3.011 μm with 540 J input to the lamps (340 J corrected for length mismatch). This was significantly more than could be obtained from a longer and higher quality laser rod of 15% Ho:YAG operated under identical conditions.

More unusual lasing properties were found when the 2.94-μm output coupler was replaced with a mirror which had a reflectivity of 90% at 1.34 μm and 88% at 1.06 μm.

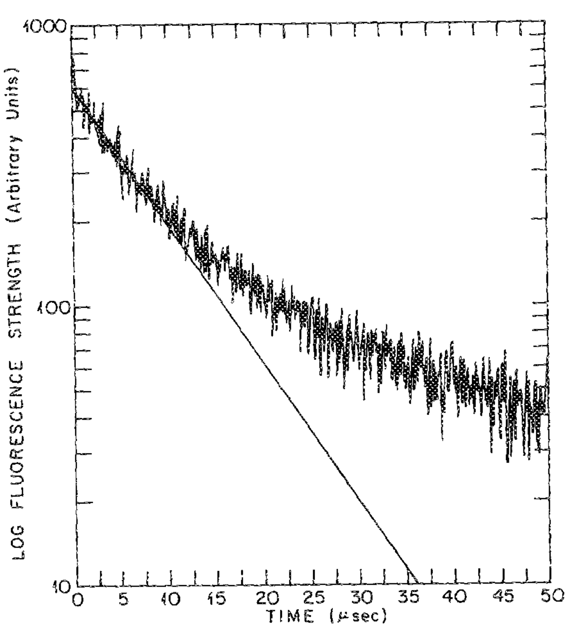

FIG. 2. Log of fluorescence signal decay vs time for the 1.06-μm emission from the Nd^{3+} $^4F_{3/2}$ level in (10% Ho, 1% Nd):YAG.

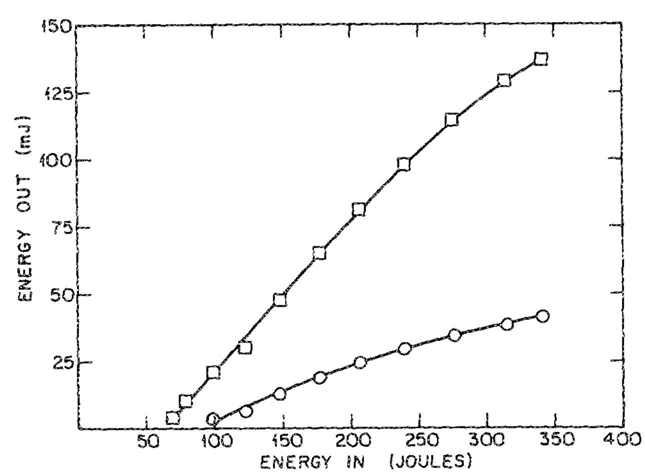

FIG. 3. Long pulse energy out vs energy in for (10% Ho, 1% Nd):YAG. The input energy plotted is the input to the flashlamps corrected for the mismatch in arc and exposed rod length. The output coupler used was designed for 90% reflectance at 2.94 μm and had a transmission of 3% at 1.06 μm. Reflectance at 1.34 μm was less than 10%. Circles are combined energy at 2.940 and 3.011 μm. Squares are output energy at 1.064 μm.

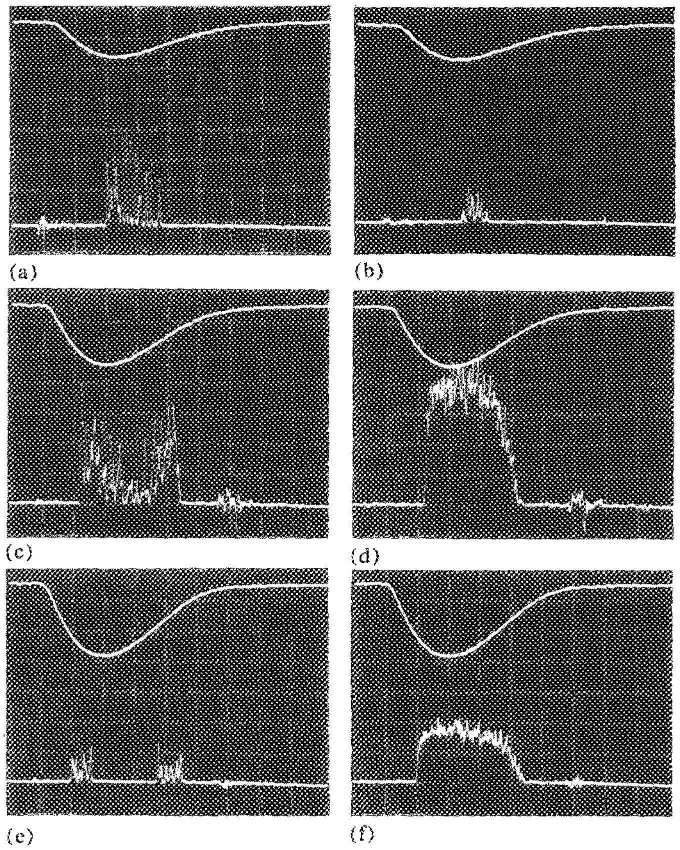

FIG. 4. Laser waveforms for the Nd^{3+} 1.339 and 1.064 μm signals from (10% Ho, 1% Nd):YAG. The upper trace monitors the current pulse to the flashlamps. The horizontal scale is 50 μs/div in all oscillographs. Waveforms at 1.339 (a) and 1.064 μm (b) with 69 J (corrected) input. Vertical scale is 10 mV/div. Waveforms at 1.339 (c) and 1.064 μm (d) with 148 J (corrected) input. Vertical scale is 20 mV/div. Waveforms at 1.339 (e) and 1.064 μm (f) with 207 J (corrected) input. Vertical scale is 50 mV/div.

This mirror combination produced simultaneous lasing at 1.064 and 1.339 μm. The unique temporal behavior of this lasing is illustrated in Fig. 4. The 1.339-μm transition has a lower threshold than the 1.064-μm [see Figs. 4(a) and 4(b)]. As the input energy is increased the 1.064-μm output rapidly becomes more intense, and the 1.339-μm lasing only occurs at the leading and trailing edges of the 1.064-μm lasing [see Figs. 4(c)–4(f)]. This may result from the residual absorption of holmium at 1.064 μm. Although small, it is apparently sufficient to alter the threshold condition for the two transitions, allowing 1.339 μm lasing to occur at low pump levels. It is possible that at higher inputs the inversion is large enough to enable 1.064 μm lasing and this transition then competes successfully with the 1.339-μm transition for the inversion.

When the total reflector was changed to one which had a maximum reflectivity at 1.34 μm and $R < 40\%$ at 1.06 μm, only 1.339 μm lasing was observed with no temporal breakup of the pulse. The maximum energy out of the rod at 1.339 μm under these conditions was 260 mJ.

In this initial work, no attempt was made to optimize any of the parameters involved to maximize output at a particular wavelength. The concentrations of the dopants can be varied, and different host crystals are being considered to improve the performance of this dopant combination. Improved material and an optimization of mirror reflectivities at the various wavelengths involved are expected to increase lasing performance. Work is in progress to understand the ion-ion interactions in this material. Such multiple wavelength lasers have potential for medical, military, and scientific applications.

Simultaneous lasing by Ho^{3+} and Nd^{3+} ions has been observed in doubly doped (Ho, Nd):YAG for the first time. Strong ion-ion interactions are indicated by the drastic changes in the Nd^{3+} $^4F_{3/2}$ and Ho^{3+} $^5I_7$ lifetimes. These interactions produce more efficient 3 μm lasing from Ho^{3+}, and some unusual lasing properties from Nd^{3+}, as 1.064 and 1.339 μm lasing occur simultaneously. With appropriate mirrors, it is believed that all four wavelengths would lase at the same time.

This work was supported by the Air Force Office of Scientific Research under contract No. AFOSR-84-0378. Cooper Laser Sonics provided a gift to support this work. C. Enderby of Cooper Laser Sonics provided impetus to this work by suggesting that dual wavelength operation might occur in (Er, Nd):YAG and would be desirable for medical applications. Valuable discussions with W. Q. Shi are also acknowledged.

[1]A. A. Kaminskii, T. I. Butaeva, A. O. Ivanov, I. V. Mochalov, A. G. Petrosyan, G. I. Rogov, and V. A. Fedorov, Sov. Tech. Phys. Lett. **2**, 308 (1976).
[2]M. Kh. Ashurov, Yu. K. Voron'ko, E. V. Zharikov, A. A. Kaminskii, V. V. Osiko, A. A. Sobol', M. I. Timoshechkin, V. A. Fedorov, and A. A. Shabaltai, Inorg. Mater. **15**, 979 (1979).
[3]A. A. Kaminskii, V. A. Fedorov, and I. V. Mochalov, Sov. Phys. Dokl. **25**, 744 (1980).
[4]A. A. Kaminskii, *Laser Crystals* (Springer, Berlin, 1981).
[5]A. A. Kaminskii, V. A. Fedorov, A. O. Ivanov, I. V. Mochalov, and L. I. Krutova, Sov. Phy. Dokl. **27**, 725 (1982).