Ho:YLF pumped HBr laser

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Abstract: A Ho:YLF laser pumped HBr molecular laser was developed that produced up to 2.5mJ of energy in the 4 micron wavelength region. The Ho:YLF laser was fiber pumped using a commercial Tm:fibre laser. The Ho:YLF laser was operated in a single longitudinal mode via injection seeding with a narrow band diode laser which in turn was locked to one of the HBr transitions. The behavior of the HBr laser was described using a rate equation mathematical model and this was solved numerically. Good agreement both qualitatively and quantitatively between the model and experimental results was obtained.

OCIS codes: (140.0140) Lasers and laser optics; (140.3070) Infrared and far-infrared lasers

References and links

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1. Introduction

Mid-IR laser sources emitting radiation in the 2 to 4 micron region are of interest due to various possible applications. Many molecules have absorption features in this wavelength range, therefore these lasers are of particular interest for molecular spectroscopy and remote sensing. An atmospheric transmission window exits both in the 2 and 4 micron regions making these lasers particularly attractive for applications that require propagation through the atmosphere. Various laser sources in this region exist such as solid state laser pumped optical parametric oscillator (OPO) systems as well as chemical lasers such as the HF/DF laser. Due to possible optical damage to the non-linear crystals OPO based systems are not really suited for scaling to multi Joule level energies. The chemical lasers are large and cumbersome to operate and use hazardous chemicals. Diode pumped solid state lasers pumping a molecular gas to produce an optically pumped molecular laser (OPML) is an approach that seeks to combine the efficiency of diode pumped solid state lasers with the scalability of gas lasers. Various such systems have been reported in the literature [1,2].

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Miller et al [1] reported a system based on pumping an HBr cell using the 2 micron output radiation of a Nd:YAG pumped OPO. They reported output energy of 0.85mJ at 4 micron. Kletecka et al [2] reported an HBr cell pumped by a 1.3 micron Nd:YAG laser. The 1.3 micron output was obtained by temperature tuning of a mode-locked Nd:YAG laser. Cascade lasing at 4 micron was reported with this system.

In this study a Tm:fibre pumped Ho:YLF laser was used to pump an HBr cell. The pump wavelength is the 2.064 micron output of the Ho:YLF laser. The high gain cross section and long upper state lifetime as well as the birefringent nature of the YLF crystal makes Ho:YLF particularly suitable for producing high energy pulses [3]. However Ho:YLF is not well suited to direct diode pumping by the widely available 0.8 to 0.85 micron laser diodes, but, can be pumped by a Tm doped laser. The availability of high power commercial Tm:fibre lasers makes a Tm:fibre pumped Ho:YLF laser an attractive candidate for producing Joule level 2 micron pulses from a relatively compact setup. The narrow absorption bandwidth of HBr necessitated the development of a frequency stabilized single longitudinal mode high energy Ho:YLF laser and this was used to pump an HBr cell.

The energy level diagram of the excitation and lasing process for an optically pumped HBr laser is shown in Fig. 1. The 2 micron pulse from the Ho:YLF laser excites the molecule from the ground vibrational level to the second excited level. Lasing at 4 micron occurs between the first and second vibrational levels. Due to the large vibrational level spacing the population of the lower laser level at room temperature is nearly zero. Therefore, after pumping by the 2 micron laser, the population inversion is essentially equivalent to the population of the V=2 vibrational level.

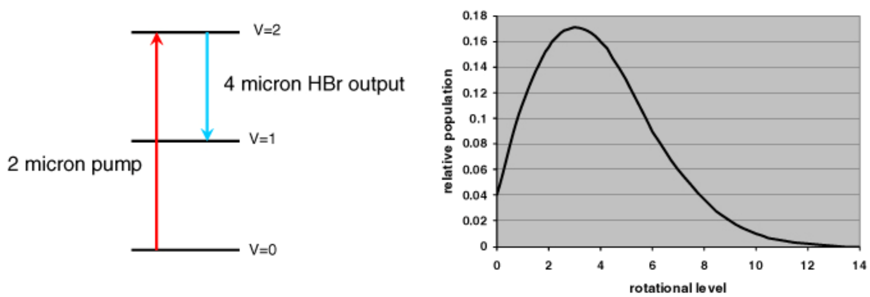

Fig. 1. The energy level diagram of a 2 micron pumped HBr laser is shown on the left. Pumping is from the ground vibrational level (V=0) to the V=2 vibrational level. Lasing occurs from V=2 to V=1. The rotational partition function of the vibrational ground state calculated at 300K is shown on the right.

A disadvantage of the Ho:YLF laser as far as pumping an HBr laser is that the output wavelength that can be produced efficiently is in the region of 2.064 micron. This corresponds to the P9 transition of HBr [1]. The calculated partition function of HBr at 300K is also shown in Fig. 1. As can be seen the relative population in the j=9 level is low compared to the j=3 level for example. Due to the relatively low population of the level that can be pumped by the output of the Ho:YLF laser it can be expected that the absorption per unit length of the HBr gas will be low. This might have an influence on the efficiency of the laser. It was not clear whether pumping on the P9 line would produce the same efficiency as that obtained by pumping on one of the levels with relatively high populations such as using the P3 transition for example. Previous results reported [1] were obtained by pumping on these relatively high population levels.

2. Experimental set-up

Figure 2 shows a schematic of the single frequency Tm:fibre pumped Ho:YLF laser as well as the HBr cell. A commercial Tm:fibre laser with a rated maximum output power of 80W (Model TLR-80-1940 from IPG Photonics) operating at nominally 1.9 micron is used to pump a Ho:YLF oscillator, the remaining pump light is used to pump a Ho:YLF pre-

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amplifier. The oscillator is operated in a ring configuration and is seeded by a single frequency distributed feedback (DFB) laser diode. See references [4] and [5] for more details regarding this laser. The 2 micron output radiation is used to pump a gas cell filled with HBr. The frequency of the seed laser is locked to one of the HBr transitions using an HBr absorption cell with a Pound-Drever-Hall feedback loop [6]. The Ho:YLF laser produced frequency stable single longitudinal mode pulses with an energy up to 70mJ per pulse. A typical spatial beam profile of the single longitudinal mode pulses produced by this laser is shown in Fig. 3. Also shown in Fig. 3 is the output energy vs. average pump power. The energy of 70mJ per pulse was obtained up to a pulse repetition rate of 50Hz. Beyond 50Hz there was a decrease in the output energy. The goal of this research project was to obtain the highest possible energy with this particular setup and hence the pulse repetition rate of the system was limited to 50Hz.

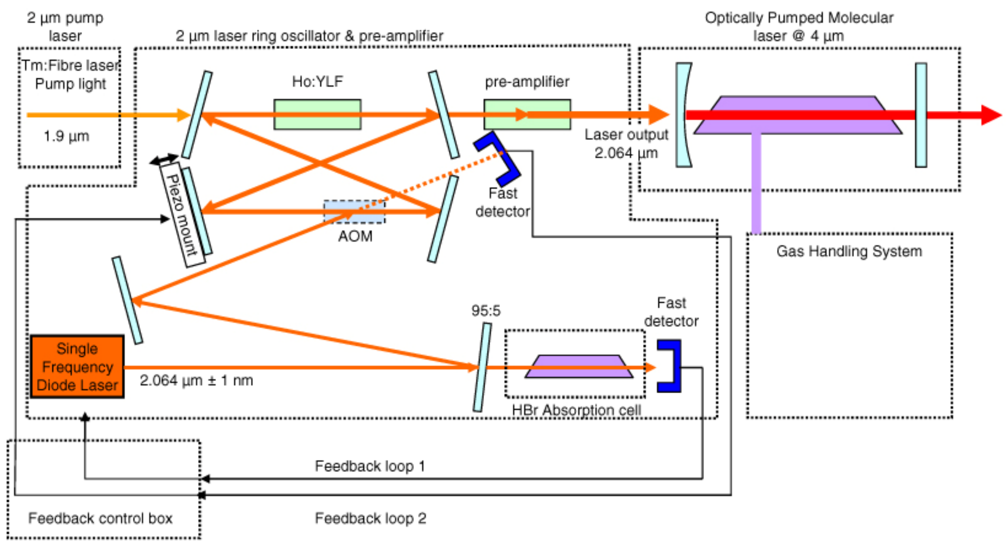

Fig. 2. Schematic outlay of the Ho:YLF pumped HBr laser

The output mirror of the HBr cell was flat with a curved input mirror with a radius of 10m. The output coupler reflectivity was 80% at 4 micron with a high reflectivity coating at 2 micron. This resulted in a double pass through the HBr cell. However, the HBr laser axis was slightly misaligned with the pump laser axis to prevent back-reflection into the Ho:YLF pump laser. This resulted in a misalignment between the two pump passes in the HBr cell. This also means that the two pump passes were misaligned with the resonator axis of the 4 micron resonator. If this is taken into account and the pump length is calculated then due to geometric considerations the pump length is equivalent to a single pass through the HBr cell. . The length of the HBr cell used in this set-up was 0.94m. The various 0 to 2 vibrational transition wavelengths and A-coefficients are given in [1]. The transition that can be accessed with this specific Ho:YLF laser is the P9 line with wavelength of 2.06412 micron. Since this is a P-transition $(\Delta j = -1)$ it is a transition from rotational levels 9 to 8

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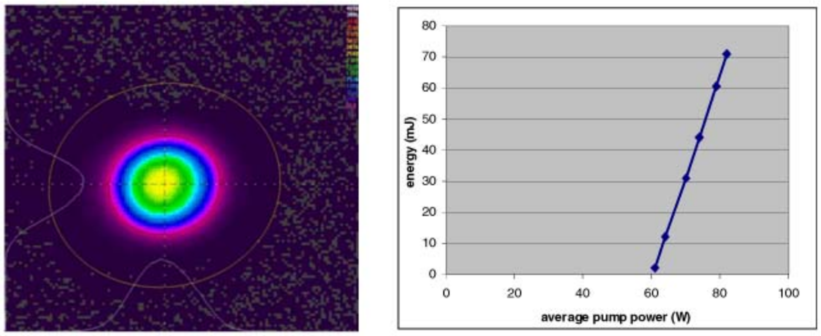

Fig. 3. The measured spatial profile of the Ho:YLF pulse is shown on the left. The output 2 micron pulse energy as function of the 1.9 micron average pump power is shown on the right.

3. Results and discussion

A multilevel numerical model, taking account the first 10 P and R branch transitions was developed for the HBr cell pumped by a nominal 2.0 micron Ho:YLF laser. The model included rotational thermalization within a specific vibrational band as well as vibrational relaxation of both the upper and lower vibrational levels partaking in the laser action. The model is similar to that reported in [2] and [7]. The pump pulse was segmented in time and each segment was propagated through the HBr cell using the Beer-Lambert law. After propagation of all time segments through the cell the output pulse is reconstructed by adding all the time segments. After the propagation of a specific segment through the laser medium the population in the various ro-vibrational levels was calculated. The intensity used in this calculation was the average value over the pump length. The average value was obtained by propagating the specific segment through the cell using the Beer-Lambert law and then the average value theorem for integrals was used to get the average value over the pump length and is given in Eq. (1).

$$I_{pump} = \left[\frac{1 - e^{-\alpha l}}{\alpha l} \right] \quad (1)$$

Here α is the absorption coefficient and l is the effective pump length. This new population, obtained by the calculation above, was then used to calculate a new absorption coefficient. Thus for a specific time segment saturation effects are not taken into account. However, the fact that new population values in the different levels are calculated after each time segment propagated through the cell meant that saturation effects are approximated for the pulse in total.

The set of coupled differential equations as given in [2] was solved simultaneously for the first 10 rotational levels of both the upper and lower vibrational levels. It was assumed that pumping occurs on a single P branch transition and specifically the P9.

Figure 4 shows the measured and calculated absorption of HBr on the P9 line as a function of pressure. This was measured and calculated at a pump energy value of 40mJ. A value of 430MHz for the Doppler broadening and 8.0MHz per torr for the pressure broadening gave the best fit to the experimental results. The pressure broadening compares well with the value of about 10 MHz per torr quoted by Miller et. al. [1] however they quoted a value of the only 205MHz for the Doppler width, while Nicholson et. al. [8] quoted a value of 400MHz for the Doppler broadening at room temperature and a pressure of 30 torr, which is close to the value that gave the best fit in this model. The flattening of the graph at higher pressures is caused by the pressure broadening of the individual HBr laser lines. The rotational thermalization time used in the model was $6.0\text{x}10^{-11}$ cm^3/molecule.s [1].

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The output energy of the laser as function of 2 micron pump energy is shown in Fig. 5. The maximum energy that was obtained was approximately 2.5mJ at a pump energy value of 60mJ, the operating pressure was 60mBar.

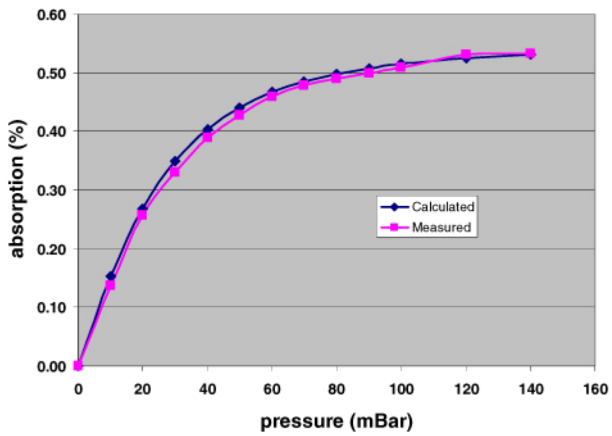

Fig. 4. Calculated and measured absorption for the P9 pump line

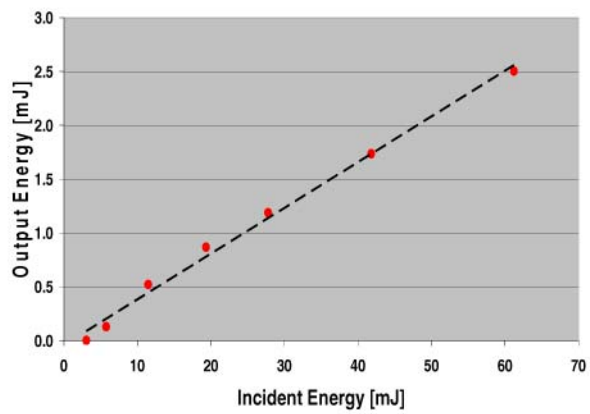

Fig. 5. Four micron output energy as function of the input pump energy

The measured time behavior of the HBr output pulse is shown in Fig. 6. The HBr cell operating pressure was 50mBar with pumping energy of 40mJ. The pump pulse duration was approximately 366ns at FWHM. The spectral component of the output pulse was measured and it consisted of mainly two lines, i.e. the P4 and P5 lines. The temporal pulse shape calculated by the numerical model using the same parameters as was used in the experiments is shown in Fig. 7. The “4 micron” in the legend on the graph refers to the spectrally integrated pulse, i.e. the sum over all ro-vibrational lines taking part in the laser action. According to the model predictions at pressures above 20 mBar the laser output consists predominantly of three lines, i.e. the P3, P4 and P5 lines with the P4 line the strongest followed by the P5, independent of the wavelength of the pumping laser. This is consistent with the experimental results obtained here and is also consistent with the results reported by Miller et al [1]. A typical spatial beam profile of the OPML is also shown in Fig. 6. It is clear that the 4 micron output beam profile is elongated in the vertical direction; this is due to the fact that laser axis was misaligned with the pumping laser axis. This was done to prevent the back reflection from entering the pump laser. The effect of this misalignment was that the geometry of the overlapping two pump beams i.e. incoming and reflected beams had to be taken into account when the efficiency of the laser with respect to absorbed energy was calculated. The experimental efficiencies shown in Fig. 8 were calculated taking these geometrical considerations into account.

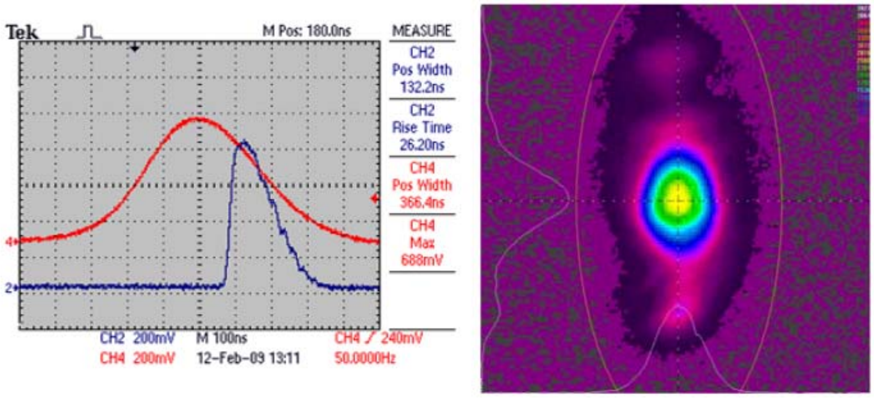

Fig. 6. The measured temporal and spatial beam profiles of the HBr laser are shown in the two figures above. On the left hand side is the temporal HBr 4 micron output pulse in blue with the 2 micron pump pulse in red. The pump duration (FWHM) was approximately 366ns and the output pulse width of the 4 micron pulse at FWHM was 132ns. The 4 micron spatial beam profile is shown on the right.

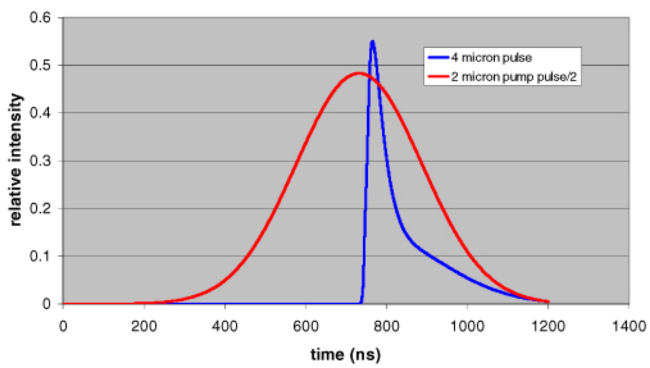

Fig. 7. Calculated temporal output pulse in blue shown with the 2 micron pump pulse in red. The FWHM of the pump pulse is 366 ns.

Figure 8 gives the measured as well as calculated efficiencies of the HBr laser relative to the absorbed pump energy as a function of the HBr laser operating pressure. The beam radius of the pumping beam was 1.5mm and that of the fundamental mode in the resonator was 1.9mm. Therefore the resonator mode was larger that the pumping laser mode and this resulted in a reduced efficiency of converting the 2 micron pump energy to 4 micron output energy. Ideally the pump laser pulse should be at least as large as the fundamental laser mode.

The efficiency of converting the 2 micron pump energy to 4 micron output energy is primarily controlled by the rotational and vibrational relaxation times. As the pressure of the laser medium is increased, the rotational and vibrational relaxation times decrease, causing a de-population of the vibrational levels which reduces the gain. The decrease in the vibrational relaxation time with increasing pressure limits the maximum operating pressure of the laser due to the loss of upper laser level population. In the case of low pressure and hence slow rotational thermalization time all the pump energy might be stored in a specific rotational level with no relaxation during the laser pulse. In this case the maximum efficiency of the laser can be expressed as [1]

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$$\frac{E_{laser}}{E_{pump}} = \frac{1}{\left(1+\frac{g_2}{g_1}\right)}\frac{\lambda_{pump}}{\lambda_{laser}} \quad (2)$$

With g_1 and g_2 the degeneracy of the lower and upper rotational levels and the $\lambda's$ the wavelength of the pump and the HBr laser. If Eq. (2) is applied to our specific case, i.e. pumping from j=9 in the ground vibrational state (V=0) to j=8 in the upper vibrational state (V=2) level with a pump wavelength of 2.06412 micron, the efficiency would be approximately 26%. A similar efficiency value will be obtained if lasing occurs on all the lines from a fully thermalized upper level [1]. However, if only a few of the strongest lines lase with the rest under threshold the expected efficiency is in the region of 14% [1]. In practice one would therefore expect a maximum efficiency of between 14% and 26% since the system might not be fully thermalized. If the pump energy is large enough to drive an appropriate level of saturation in the pump transition then a cascade lasing process [2] including vibrational level 1 to ground is predicted. In this case Eq. (2) is no longer valid and significantly higher efficiencies might be possible.

The efficiency of approximately 12% obtained in this experiment is thus clearly significantly lower that the maximum that can be expected from a 2 micron pumped HBr laser. The measured efficiencies as a function of absorbed pump energy are shown in Fig. 8. As can be seen, there is a reduction in efficiency at higher pressures due primarily to the vibrational de-activation of the upper laser level. The efficiency calculated for this specific set-up, taking the pumping geometry into account is also shown is Fig. 8. The experimentally measured efficiency, as function of pressure, decreases faster that that predicted by the model. The reason for this is not clear. A faster vibrational relaxation constant will explain the decrease. In this model the values published in [1] was used. It is important to mention that the fastest v-v relaxation constant used here is a value of $1.5\text{x}10^{-11}$ cm^3/molecule.s. This is for v-v exchange between the two HBr isotopes. The other v-v relaxation rates are an order of magnitude less than this value. The significance of this is the fact that an HBr laser operating on a single isotope will have significantly less pressure dependence that the dual isotope case. Another possible cause for the difference between the predicted and measured efficiencies could be pressure enhanced non-resonant absorption by the other HBr isotope. Also shown in Fig. 8. are "optimum" calculated efficiencies for this particular (i.e. P9 pumped) case. As can be seen the maximum obtained from the model is approximately 20% which compares well with the theoretical (non-cascade lasing) value of 26%. In this optimization the geometry of the pump laser was chosen to overlap completely with the fundamental laser mode, in addition the output coupler value used was 60% and the pumping energy was 100mJ. It is possible that a multi-parameter optimization of the system will give values closer to that predicted by Eq. (1). In addition, pumping the system with a much higher energy will result in the saturation of the pump transition leading to cascade lasing. This will have a positive effect on the efficiency of the system.

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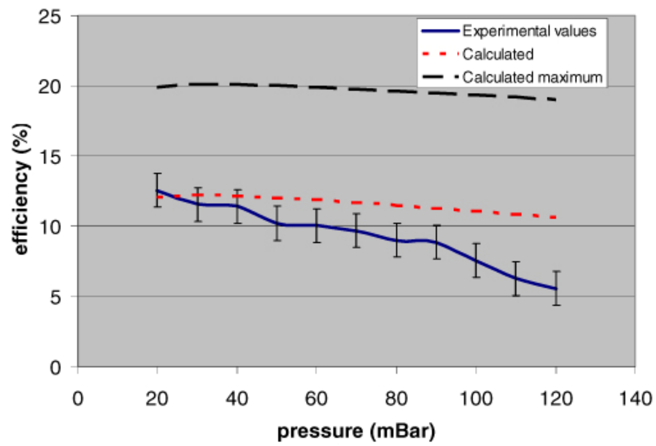

Fig. 8. Measured and calculated efficiencies of converting 2 micron pump pulses to 4 micron output pulses. Efficiency is expressed relative to the absorbed pump power.

4. Conclusion

A Ho:YLF pumped HBr laser emitting radiation in the 4 micron wavelength region was demonstrated. The laser was pumped on the P9 (2.06412 micron) line of HBr. At pressures above 20 mBar lasing was observed on the P4 and P5 lines of HBr. This indicates that thermalization of the rotational lines occur during the laser pulse. A maximum efficiency of approximately 12% of converting absorbed 2 micron pump photons to 4 micron output pulses was obtained. A numerical model was used to predict the expected efficiency of this particular setup. The efficiency predicted by the model was slightly higher than the measured values particularly at higher pressure values. The model was also used to optimize the expected 4 micron output energy of this setup. Optimum efficiency is predicted if the pump and resonator axis are properly aligned, and when the pump and resonator modes are properly matched. In addition the output coupler reflectivity was optimized using the numerical model. The predicted efficiency of such an optimized system is approximately 20%. Because relatively high conversion efficiencies of 2 micron radiation (derived from one of the strong Ho:YLF lines) can be obtained and the possibility that a Ho:YLF oscillator/amplifier system can be scaled to Joule level energies makes this system an attractive candidate for producing Joule level 4 micron pulses.

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