Mode-locked picosecond diamond Raman laser

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We present a mode-locked diamond Raman laser synchronously pumped by a mode-locked laser running at 532 nm and pulse duration 26 ps. The diamond laser generated up to 2.2 W of average power with output pulses of duration 21 ps at a yellow wavelength of 573 nm. The output pulse duration varied notably with small changes in cavity length and decreased to a minimum of 9 ps. The power and pulse duration behavior as a function of cavity length is explained well by a model that includes phonon dephasing and group velocity dispersion of the pump and Stokes fields. © 2010 Optical Society of America

Diamond has properties that make it highly attractive as a laser material, such as its exceptional thermal conductivity, wide band gap, and lack of first-order IR absorption bands. Considerable efforts over the past two decades to realize semiconductor lasers [1], color center lasers [2], and rare-earth doped lasers [3] based on doped diamond have not yet led to practical devices. On the other hand, advances in the synthesis of high purity and low-defect single crystal diamond [4] have recently enabled the development of efficient diamond Raman lasers operating in the nanosecond regime taking advantage of its high Raman gain coefficient [5]. The range of performance capabilities envisaged for diamond Raman lasers is essentially untapped, including operation in the transient picosecond regime.

Picosecond Raman lasers are in demand for a range of nonlinear optical applications that require specific output wavelengths, such as yellow (550–600 nm) probe pulses for biological imaging using two-photon fluorescence [6]. Raman lasers also potentially allow generation of multiple Stokes wavelengths simultaneously or at a selected Stokes order according to the Raman resonator design [7]. Though picosecond Stokes generation within one or two passes of the Raman medium can be efficient [8], the pulse-power threshold is much higher than for Raman oscillators, the output spectrum is not easily controlled, and the output beam is not of sufficient quality to meet the demands of most applications. Synchronously pumped Raman resonators provide a method to realize low threshold and efficient picosecond Raman lasers at the first Stokes and with excellent beam quality [9,10]. Synchronously pumped lasers rely on matching the interpulse period of the pump laser with the round-trip time of the Raman laser resonator to build up an intense circulating picosecond pulse in the Raman resonator over many pulses. Using a 50-mm-long KGd(WO₄)₂ (KGW) crystal pumped by a 2 W 532 nm pump laser of pulse duration 10 ps, we observed 25.6% conversion to 559 nm and shortening of the pulse durations to 3 ps by detuning of the Raman resonator cavity length. Using diamond potentially offers a greatly extended range of capability. The larger Stokes shift (1332 cm⁻¹) compared to KGW (768 and 901 cm⁻¹) enables an output wavelength of 573 nm from a single Stokes shift when using a 532 nm pump laser. Diamond has a much higher gain coefficient [11,12], enabling smaller crystals to be used. The longer dephasing time of diamond (6.8 ps [13]) compared to 3.2 ps for KGW is expected to place a higher limit on the pulse duration and will enable us to test models for pulse compression limits in synchronously pumped Raman lasers. Also, the outstanding thermal conductivity of diamond will allow rapid heat removal and thus potentially very high average output powers.

In this Letter we present a synchronously pumped diamond Raman laser generating up to 2.2 W at the 573 nm first-Stokes wavelength, using a 6.7-mm-long diamond crystal. We present a numerical model of the system that assists in explaining the observed laser conversion efficiency and pulse duration as a function of the cavity length.

The diamond laser cavity was a z-fold configuration comprised of two curved mirrors (M₁ and M₂, with concave radii of 200 mm) and two plane mirrors (M₃ and M₄) as shown in Fig. 1. The fold angle was set to 6° to compensate for the astigmatism introduced by the 6.7 mm Brewster-cut diamond crystal. The mode locked Nd:YAG laser was frequency doubled to 532 nm and focused through M₁ into the diamond crystal with lens L1 to approximately match the 32 μm (1/e² radius) cavity mode waist. Up to 7.5 W at 532 nm was incident on the diamond crystal, with the pulse train composed of 26 ps pulses at a

![Fig. 1. (Color online) Experimental layout of the synchronously pumped Raman laser.](image-url)
repetition rate of 78 MHz. Mirrors $M_1$, $M_2$, and $M_3$ were highly reflecting at the first-Stokes wavelength of 573 nm, and the output coupler $M_4$ had a transmission of 12%.

The position $\Delta x$ of mirror $M_4$ was tuned to optimize the performance of the laser; $\Delta x=0$ is defined as the cavity length measured to have the lowest threshold, and negative values correspond to a shortened cavity. We first optimized the laser for maximum output power, which was achieved for $\Delta x=+50 \mu m$. For a pump power of 7.5 W, we measured an output power of 2.21 W at 573 nm. The laser threshold was approximately 2 W, leading to a slope efficiency of 41% and an absolute efficiency of 29%.

Figure 2 shows the power and pulse duration of the output at 573 nm as a function of $\Delta x$, measured for an input pump power of 7 W. The pulse duration was measured with a scanning second-harmonic-generation autocorrelator, with the pulse durations inferred, assuming that the pulses were Gaussian in time. The output power behaved differently for positive and negative $\Delta x$; substantial negative detunings of up to $\Delta x=-900 \mu m$ were tolerated (which corresponds to a temporal mismatch of 6 ps per round trip), whereas a positive detuning of just $+200 \mu m$ (1.3 ps) caused the laser action to cease. The highest output power was observed for a detuning of $+50 \mu m$, for which the output pulse duration was 21.3 ps (cf. pump pulse duration 26 ps). For shorter cavity lengths, the pulse duration monotonically increased up to a maximum of 30 ps for $\Delta x=-750 \mu m$, while the output power decreased by $\sim 50\%$. For longer cavity lengths ($\Delta x=+200 \mu m$), there was a sharp reduction in pulse duration down to 9 ps; however, the output power also decreased rapidly so that no enhancement in peak power was observed in this pulse-shortened regime.

The capability to shorten and compress pulses is important for many applications, and thus we have developed a numerical model to analyze the factors that influence pulse duration and peak power. The model uses the equations for transient Raman scattering, tracking the amplitudes of the Stokes and pump pulses as well as the phonon excitation, and also accounts for group velocity walk-off between the pump and Stokes pulses through the crystal. These equations are given for example in Penzkofer et al. [14] (Eqs. 77–79), using different velocities for the Stokes and pump pulses, and with the assumption that the excess population of phonons is small. After using the finite difference method to transform the time- and space-dependent equations into a first-order-accurate set of time-dependent equations on a spatial grid, we solve the equations numerically using a Runge–Kutta algorithm. We solve for a sequence of single passes through the crystal, using the output Stokes field from one pass as the input Stokes field for the following pass; the simulation is terminated when the Stokes pulse has reached its steady-state profile. We solve the equations in a frame moving at the Stokes group velocity, in order to avoid numerical dispersion for the resonated Stokes field. The cavity length detuning is simulated by retarding or advancing the Stokes pulse before it is recycled after each round trip. We used the experimentally known parameters for the pump power (7 W), duration (26 ps, assuming a Gaussian temporal profile), cavity mode waist (31 $\mu m$), output coupling (12%), and diamond length (6.7 mm). We used a phonon dephasing time of 6.8 ps [13]. The Raman gain and residual resonator losses were adjusted in the model to achieve best fit with the experimental data. The chosen Raman gain at 532 nm, which is not well known, was 50 cm/GW, consistent with that expected from measurements performed at other wavelengths [11,12]. The best fit was obtained for additional resonator losses of 13%, which is reasonable considering our measured mirror leakages and including some other minor losses including scatter and reflection losses from the Brewster facets.

The model results for the output power and pulse duration (calculated from the output pulse profiles by simulating an autocorrelation measurement to allow comparison to the experimental values) are included in Fig. 2. The close fit with the experimental data obtained for both variables as a function of $\Delta x$ indicates that all the key physical processes are included in the model.

To elucidate the pulse-shortening mechanism, we analyzed the pulse formation as a function of cavity length. In Fig. 3 the steady-state pulse shapes of the pump and Stokes pulses both before and after a transit through the diamond crystal are shown for three different regimes: near maximum efficiency ($\Delta x=+60 \mu m$), strong pulse shortening ($\Delta x=+180 \mu m$), and the condition for longest output pulses ($\Delta x=-900 \mu m$). The zero of the time axis is defined such that a Stokes pulse moving through the crystal at its group velocity will not shift in time.

For the cavity length corresponding to the maximum power output ($\Delta x=+60 \mu m$; central pair of plots in Fig. 3), the Stokes and pump pulses are well over-

![Fig. 2.](https://example.com/fig2.png) (Color online) Power output (open squares) and pulse duration (filled squares) of the 573 nm laser output as a function of cavity length. Also shown are the results of the numerical simulation (power—solid line, duration—dashed line).
The model shows that the important factor for simultaneous efficient operation and pulse compression is a large temporal walk-off between the Stokes and pump pulses during each transit through the crystal, which allows the shorter Stokes pulse to walk through and extract the energy from the entire pump pulse. The required walk-off is more easily achieved for short pump pulses and for long Raman media with a high group velocity dispersion. The compression will thus be larger for longer lengths of the diamond crystal and for materials with higher dispersion. This is consistent with the larger pulse compression observed for the KGW Raman laser of [10]. In that work, the crystal was 7 times longer, and the pump pulse was 2.6 times shorter than the present work (the group velocity mismatch is similar) [10]. The compression effect will be examined in more detail in future work along with more details of the modeling method.

In summary, we have demonstrated a diamond Raman laser synchronously pumped at 532 nm by a mode locked Nd:YAG laser generating 2.2 W at 573 nm. Our numerical model of transient Raman scattering explains the extreme asymmetry of the laser power and pulse duration with cavity length detuning as a consequence of operating in the transient Raman scattering regime. These sources offer an efficient and robust method of generating ultrashort pulses at a range of hard-to-reach wavelengths in the yellow and orange spectral regions from industry-standard picosecond lasers, with many important applications such as biological imaging.

References