Investigating diamond Raman lasers at the 100 W level using quasi-continuous-wave pumping

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Quasi-cw pumping is used to investigate the high-power characteristics of cw beam conversion in diamond Raman lasers (DRLs). We show that thermal gradients establish in DRLs at approximately 50 μs for a 100 μm pump beam diameter, and thus that the steady state for cw operation can be reached within the 100–300 μs pulse duration of conventional quasi-cw pump laser technology. Using this approach, a steady-state on-time output power of 108 W was obtained from an external-cavity DRL during 250 μs pulses with 34% conversion efficiency. No thermal lens in the diamond was evident, showing excellent prospects for further power scaling.

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Diamond Raman lasers (DRLs) are rapidly emerging as an important technology for efficient high-power beam conversion [1–4]. Diamond offers extremely broad transparency ($\lambda > 230$ nm), high Raman gain (~10 cm/GW at 1 μm [4–6]) and a large Raman shift (1332 cm^{-1}) [7]; but most important are its unmatched thermomechanical properties. Although it has only become available recently as a Raman laser material due to developments in the synthesis of optical-grade, low-birefringence material, diamond has already enabled record crystal Raman laser output powers in both pulsed and cw regimes [1,2,6]. The broad wavelength coverage of diamond has been demonstrated with Raman lasers spanning the deep UV [8], visible [9], near-infrared [3,10,11], and mid-infrared [12] wavelengths.

Among diamond's many attractive properties for Raman beam conversion, a distinguishing feature is its thermal conductivity (2200 $W/m \cdot K$), which is 2 to 3 orders of magnitude higher than conventional Raman crystals. Together with its relatively low coefficient of thermal expansion ($1.1 \times 10^{-6}\ K^{-1}$) and high damage threshold, this property makes it an extremely interesting material for scaling the power of crystalline Raman lasers. To date, output powers up to 25 and 15 W have been demonstrated in pulsed [2,13] and cw DRLs [1,6,14]. These powers exceed those obtained in other crystals in which thermal effects [15,16] or, in the case of silicon, multiphoton absorption, make scaling to higher powers much more challenging [17,18]. In many of these cases, the DRL output beam quality was near diffraction-limited [1,6,13] and conversion efficiencies were above several tens of percent [1,3,14].

Our primary interest in this work is the wavelength conversion of cw beams at greatly elevated power levels. Continuous-wave operation is important from the point of view of applications and for investigating Raman laser dynamics at high average powers. Recently, cw Raman conversion was demonstrated in external-cavity DRLs with a maximum output power of 15 W [1,14]. No thermal effects were evident, and no damage was observed despite power-density in the diamond and on its antireflection (AR)-coated end facets of 43 MW/cm^2 [1]. However, the limits for power scaling in DRLs are not well understood.

The nature of the Raman interaction places some key requirements on the pump source linewidth and brightness. For low threshold and efficient conversion the pump laser should have high brightness and narrow linewidth (<40 GHz; i.e., <0.15 nm at 1064 nm) [19]. Although Yb fiber lasers readily deliver high brightness, they typically operate over a much broader bandwidth (>1 nm). Conversely, Nd:YAG lasers offer intrinsically narrow linewidth, yet achieving multiple hundred-watts of cw output with near-perfect beam quality from such lasers is a significant challenge [20].

Here we report a novel approach for investigating power scaling of external-cavity DRLs with relaxed pump requirements, enabled by the rapid thermal dissipation rate of diamond. We show that the factors limiting performance at high power are either those that occur over very short time-scales (such as optical damage) or that are due to the establishment of thermal gradients in the crystal. Steady-state output is found to be attained well within the typical 100–300 μs pulse duration of quasi-cw lasers, enabling high-power conversion to be investigated using much simpler pump laser technology. By employing readily available quasi-cw-pumped Nd:YAG technology we used 320 W of pump power to generate more than 100 W on-time Stokes output for durations of 200 μs. The DRL operated with 34% conversion efficiency and excellent beam quality, and showed no evidence of thermal effects or damage to the diamond even at the highest power.

For external-cavity cw DRLs, the factors most likely to limit output power are either catastrophic damage to the diamond or optical coatings in the cavity, or else thermal effects such as stress fracture, stress birefringence, or thermal lensing. Catastrophic damage to the bulk diamond or to intracavity optical coatings is largely dependent on peak intensities in the cavity and thus will be determined within microseconds of cw operation, whereas thermal effects in the bulk typically evolve over longer time-scales. In terms of the average heat load in the crystal, which can lead to stress fracture [21], the high

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thermal conductivity and low thermal expansion coefficient of diamond indicate that it should, in principle, be able to tolerate heat loads approaching 1 MW [22]. Thermal lensing (due to localized stresses and the thermo-optic effect) and stress-induced birefringence, however, are dependent on the thermal gradients in the vicinity of the pump beam; therefore, any thermal lens or stress-induced birefringence will be established at the same time as the thermal gradients. The temporal evolution of a thermal lens in a high average-power external-cavity Raman laser has recently been observed using potassium–gadolinium–tungstate (KGW) as the Raman gain material [23]. In that work it was demonstrated that the evolution in time of the output power and beam profile closely matched the buildup time of the thermal gradients within the pumped region of the crystal, which occurred over approximately 30 ms, corresponding to a few times the thermal time constant for KGW with a 105 μm pump beam radius (~7 ms). After this time, the output power and beam profile of the KGW laser was stable. In diamond, the thermal conductivity is three orders of magnitude higher than in KGW, and so the time constant for establishment of thermal gradients, which can be approximated as [24]

$$\tau = \frac{w^2 \rho C_p}{\kappa} \qquad (1)$$

is correspondingly much shorter than in KGW (where w = pump beam radius, ρ = material density, C_p = specific heat capacity, and κ = thermal conductivity). For a 50 μm radius pump beam, and using the values for diamond $\kappa = 2200\ \mathrm{W/m \cdot K}$, $C_p = 0.519\ \mathrm{J/g \cdot K}$, and $\rho = 3.51\ \mathrm{g/cm^3}$, the approximated thermal time constant is 2 μs. In order to illustrate the evolution of the thermal gradients in a DRL and compare them to a conventional Raman crystal (KGW), numerical simulations were performed using a finite-element-analysis transient heat solver in two dimensions (QuickField; Tera Analysis Ltd.). For simplicity, a constant beam radius through the crystal is assumed, which is reasonable in our case since the crystal length is less than the beam confocal parameter. Figure 1 shows the calculated evolution in time of the maximum thermal gradient in the crystals (normalized to the steady-state thermal gradient) for a constant heat source with Gaussian profile and radius w = 50 μm. The thermal gradients reach 99% of their steady-state values after 53 μs in diamond and 27 ms in KGW. This value for KGW is in close agreement with the experimentally observed evolution of thermal effects in the KGW external-cavity Raman laser mentioned above (32 ms) [23]. Therefore, gradient-driven thermal effects in an external-cavity DRL with a 50 μm radius pump beam (or smaller) are established and stable after a period of approximately 50 μs. In our experiments, the Raman laser performance after the initial 50 μs of each pulse is considered as cw operation for the purposes of investigating thermal effects and power scalability in the DRL. Although the peak temperature on-axis increases beyond this period, the temperature rise is not expected to significantly affect Raman conversion, as temperature-dependent effects on Raman line-shape and gain

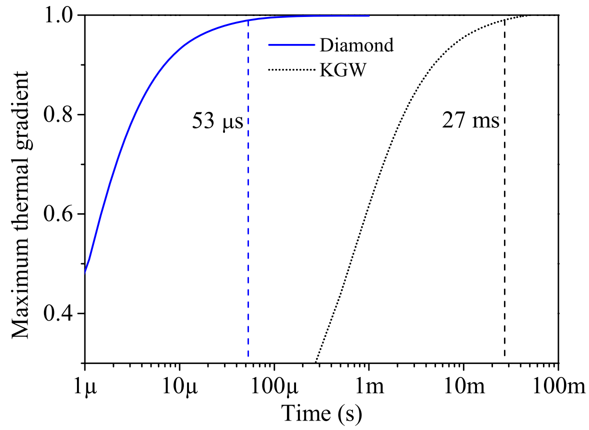

Fig. 1. Temporal evolution of maximum thermal gradients in diamond and KGW lasers under pumping with a 50 μm radius cw beam. The ordinate axis is normalized to the steady-state value in each case. The dashed lines mark the time to reach 99% of the steady-state value.

coefficient are weak for temperature increases below several hundred Kelvin [7].

Figure 2 illustrates the layout of the DRL and the quasi-cw pump. The DRL consisted of a 8 mm × 4 mm × 1.2 mm, low-nitrogen, low-birefringence, CVD-grown single-crystal diamond (ElementSix, Ltd.), placed in a near-concentric cavity designed to closely match the waist radii of the Stokes mode and the pump beam in the diamond. The concave input coupling mirror had a 75 mm radius of curvature, transmission >96% at the pump wavelength (1064 nm) and reflectivity >99.9% at the first Stokes wavelength (1240 nm). The pump beam was focused into the diamond crystal using a f = 100 mm lens with polarization aligned to the diamond ⟨111⟩ axis for maximum gain [4]. The concave output coupling mirror had a 50 mm radius of curvature, was highly reflective at 1064 nm to provide a double-pass of the pump through the diamond, and had 0.5% output coupling at 1240 nm. The diamond end facets were AR coated for 1240 nm to minimize intracavity losses.

The pump laser consisted of two Nd:YAG gain modules, side-pumped with quasi-cw diodes (RBA20–1P200, Northrop Grumman), arranged in a linear cavity with a

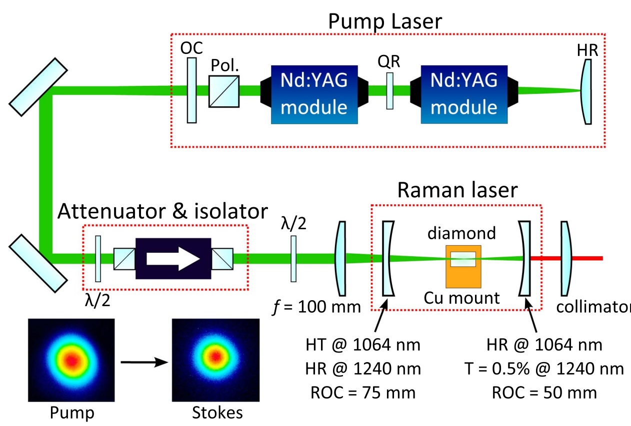

Fig. 2. Illustration of the DRL and the pump laser. Shown in the bottom-left corner are the far-field profiles of the pump (left) and Stokes beams (right). OC, output-coupler; HR, high-reflector; HT, highly transmissive.

linear polarizer and birefringence compensation provided by a quartz rotator (QR). This laser produced 250 μs pulses at 40 Hz with 370 W on-time power in a linearly polarized beam with $M^2 < 1.5$. The pump laser output exhibits very strong relaxation oscillations during the first 30 μs of each pulse but is thereafter stable with less than 1.5% noise for the remainder of the pulse (see Fig. 4).

The DRL output power and efficiency is shown in Fig. 3. The threshold was 57 W, beyond which the Stokes output power approached a slope efficiency of 34% and total conversion efficiency of 34%. The maximum on-time output power was 108 W, achieved with 322 W of pump power. The nonlinear slope efficiency observed here, which converges to the overall conversion efficiency at powers well above threshold, is due to the improved depletion of the pump beam with increased intracavity Stokes power. This characteristic of cw external-cavity Raman lasers, which is not typically observed in pulsed systems due to temporal dynamics [4], will be elucidated in a forthcoming publication. The DRL showed no sign of power roll-off or reduction in slope efficiency at the maximum power, indicating that any thermal effects in the DRL are not limiting the laser performance. The pump power in these experiments was restricted to this level in order to avoid damage to the DRL cavity mirrors, which was observed on the input coupler during prior experiments at similar power levels. The intracavity power and power-density of this laser are comparable to the highest levels for any cw solid-state laser, including kW fiber amplifiers and thin-disk lasers [25,26]. With output-coupling of 0.5%, steady-state output power of 108 W and a Stokes-mode radius of 55 μm throughout the diamond, the intracavity power exceeds 21 kW and the power density in the diamond and on its AR-coated end facets exceeds 220 MW/cm^2, which is far beyond the long-pulse/cw damage threshold for dielectric coatings on conventional substrates. This greatly increased damage threshold is likely due to the diamond acting as a very efficient heat-sink for the optical coating, thus inhibiting the thermal damage that typically causes coating failure under high-power cw operation. While the conversion efficiency of this laser is on par with previous results for external-cavity cw Raman lasers [1,14], it is well below the quantum limit (85.8%). The reduced efficiency can largely be attributed to the low output coupling of the cavity (0.5%) relative to the loss in the diamond (approximately 0.6% per round trip), resulting in less than half of the generated Stokes power being out-coupled from the cavity. The output of the laser was horizontally polarized, despite expectation from theory that the Stokes polarization should be parallel to the ⟨111⟩ crystal axis for our pumping configuration [4]. We have observed this effect in other cw DRLs and believe it is due to birefringence in the diamond, compounded by the high-Q cavity.

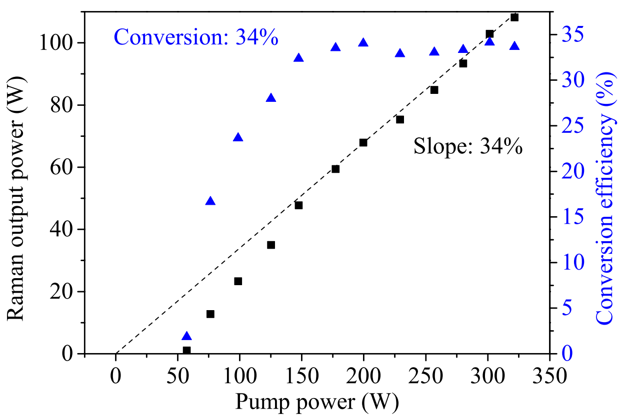

Fig. 3. Output power and conversion efficiency of the DRL.

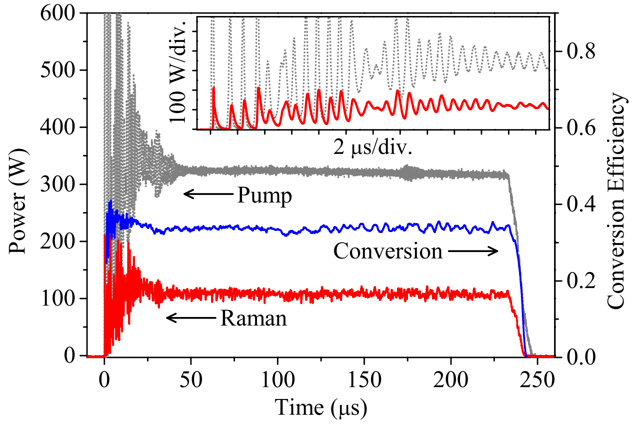

Fig. 4. Power-calibrated oscilloscope trace showing a pump pulse (grey dotted curve) and corresponding Raman pulse (red solid curve) at 108 W Stokes output power, and a moving average of the conversion efficiency (2.5 μs period, blue solid curve). The inset shows the leading edge of these pulses on an expanded time-scale.

Figure 4 shows a pump pulse and the corresponding output pulse of the DRL operating at 108 W output power, as well as a moving average of the conversion efficiency (2.5 μs period). The inset shows the leading 25 μs of this pulse on an expanded time-scale. The relaxation oscillations in the pump during the first 30 μs of the pulse are mirrored in the Raman output with some smoothing due to the long lifetime of the DRL cavity. The conversion efficiency shows a drop of 4% over the initial 35 μs of the pulse, followed by a stable level of 34%. This small initial drop could be due to a variety of factors, including settling of the spatiotemporal pump dynamics and thermal effects in the diamond. Nevertheless, the sum total of these effects is only a slight perturbation of the conversion efficiency. The output beam profile was unchanged across the full range of power investigated (see Fig. 2 for the beam profile at full power), and had an $M^2 < 1.1$, as typically observed for DRLs of lower power [1,13].

The absence of any significant thermal effects in the diamond, as well as the demonstrated ability of the diamond bulk and AR-coated facets to withstand extremely high steady-state intracavity power densities, suggests substantially higher powers are achievable with cw DRLs. The slope efficiency well above threshold was low compared to the quantum efficiency, which is attributed to substantial round-trip Stokes loss in the diamond relative to the cavity output-coupling. We therefore anticipate that higher output-coupling will provide improved overall conversion efficiency by increasing the ratio of the output-coupling to the intracavity loss, provided the pump power is sufficient to operate several times above threshold. Increased output coupling also reduces the risk of coating failure and damage to the bulk diamond, for a given output power, by reducing the Stokes field in the cavity. In future experiments we plan to investigate the DRL performance under increasing pulse duration and pump power using narrow-linewidth fiber lasers, as well as raising the efficiency using higher cavity out-coupling.

In conclusion, we have shown that quasi-cw pumping with submillisecond pulses is a convenient and effective method for investigating cw beam conversion in DRLs at high powers. Using this approach we have demonstrated 108 W steady-state Stokes output power for durations of 200 μs in an external-cavity quasi-cw-pumped DRL, which is an order of magnitude above previous cw and quasi-cw crystalline Raman lasers. Furthermore, the observed stable operation of the DRL without any evidence of significant thermal effects, and under extreme intracavity powers and power densities, shows excellent prospects for further power scaling with increased pump power and output-coupling. These results demonstrate the capability for diamond to provide cw beam conversion at output power levels above 100 W. This work also highlights the potential application of DRLs as compact external frequency converters for high-power laser platforms such as narrow-linewidth fiber lasers and thin-disk lasers.

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