# Raman Amplification of Laser Pulses in Microcapillary Plasmas

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**Abstract.** Raman amplification of ultrashort pulses is demonstrated in microcapillary plasmas. Experiments in very short microcapillaries (0.2 - 0.5 mm) with a broadband seed pulse show that the amplification factor is in agreement with the linear growth rate.

### **INTRODUCTION**

Raman amplification is based on the three-wave interaction between two counterpropagating electromagnetic waves and a plasma wave, whose frequencies and wave vectors satisfy the energy and momentum conservation relations:

$$\omega_1 = \omega_2 + \omega_n, \quad k_1 = k_2 + k_n. \tag{1}$$

where  $\omega_1$ ,  $\omega_2$ ,  $\omega_p$  are the frequencies of the pumping pulse, seed pulse, and plasma wave, respectively, and  $k_1$ ,  $k_2$ ,  $k_p$  are the corresponding wave vectors. The idea of using the Raman instability as a means to amplify and compress laser pulses has been the object of study for considerable time [1]-[3]. Early work on the compression of laser pulses in gases was reported in [1], and recently, Raman compression from tens of nanoseconds to tens of picoseconds has been achieved in gas mixtures [2]. The maximum power of laser pulses that can be compressed in gases is limited by ionization. The advantage of using plasma as the medium is that there is no theoretical limit on the laser power since plasma can handle ultrahigh power without any "damage" to itself.

In the linear regime of Raman amplification, the intensities of the pump and the seed pulses are moderate and there is no significant pump depletion. The seed pulse grows exponentially with a growth rate,  $\gamma_{RBS}$ , independent of the seed intensity [4]:

$$\gamma_{RRS} = a_1 (\omega_1 \omega_n / 4)^{1/2}$$
<sup>(2)</sup>

where  $a_1$  is the normalized vector potential of the pump;  $a_1 = 0.85 \times 10^{-9} \lambda_1 I_1^{1/2}$  (the pump wavelength  $\lambda_1$  is in µm and the intensity  $I_1$  is in W/cm<sup>2</sup>). Recently, new effects were identified in the nonlinear behavior of plasma when interacting with ultra-intense laser pulses [5]-[7]. Theoretical studies have shown that the amplified pulse duration decreases inversely proportional to the pulse amplitude and a compression from picoseconds to femtoseconds is achievable in the nonlinear regime [8]. The Raman amplification in combination with the compression effect in plasma provides a potential of overcoming the power limit of current chirped-pulse-amplification (CPA)

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techniques, set by the thermal damage threshold of the optics. Such Raman amplifiers can be useful to produce ultra-intense laser pulses for pumping soft x-ray lasers.

## **EXPERIMENTAL RESULTS**

Our experiments were initially designed for the linear regime. In this paper we present the results of Raman amplification in microcapillary plasmas. The analysis of the data suggests that the observed amplification in the initial experiment was obtained from a plasma with a length much shorter than the microcapillary length [9]. The second experiment was performed in much shorter microcapillaries to check this hypothesis, and the results show that the amplification agrees well with the estimated linear growth rate.

## Experiment with a 745 nm, 200 fs Seed Pulse in LiF Microcapillaries

The experimental setup with a 745 nm, 200 fs seed pulse is shown in Fig. 1. The pumping pulse was provided by the second harmonic of a Nd:YAG laser at  $\lambda = 532$  nm with 150 mJ in 5 ns (FWHM). The seed pulse and the pumping pulse were both focused onto the center of the microcapillary by an F/25 lens L<sub>1</sub> and an F/8 lens L<sub>2</sub>, respectively. The initial plasma was created inside the LiF microcapillary through wall ablation by a low power nanosecond KrF laser (50-60 mJ in 20 ns at  $\lambda = 248$  nm). The amplified seed pulse, propagating in the opposite direction to the pumping pulse, passed through mirrors M<sub>2</sub> and M<sub>3</sub> and was focused by a lens L<sub>4</sub> onto the entrance slit of a spectrometer with a photomultiplier detector.



FIGURE 1. Experimental setup for Raman amplification with a 745 nm seed in a LiF microcapillary.



**FIGURE 2.** Amplification as a function of the microcapillary length (a) and initial intensity of the seed pulse (b).

Figure 2 (a) shows the amplification ratio V<sub>0</sub>/V<sub>i</sub> vs. the microcapillary length at a delay of 60 ns. Each point in Fig. 2 (a) is an average of 20 shots, with the error bar calculated from the square root of standard variance. The results were obtained in two series of measurements (Exp. 1 and Exp. 2). The relatively large error was mainly due to the irreproducibility of the plasma and the intensity fluctuations ( $\sim 10\%$ ) of the seed and the pumping pulses. These results show the amplification increasing gradually with the microcapillary length. A ratio  $V_0/V_1$  of ~ 5 - 6 was reached inside the  $L_{cap} = 3$ mm microcapillary. However, the increment of the amplification as a function of the microcapillary length is much less than the exponential growth predicted by the linear theory. The intensity of the seed pulse was also varied by inserting filters between  $L_1$ and the chamber window. The amplification ratio for three initial seed pulse intensities, 0.6, 1.2, and  $8.5 \times 10^{12}$  W/cm<sup>2</sup> (corresponding energies: 2, 4 and 30 µJ) in a 2 mm microcapillary are shown in Fig. 2 (b). It was found that the amplification ratio drops as the intensity of the seed pulse  $(I_2)$  approaches the intensity of the pumping pulse (I1). When I2 was significantly smaller than I1, the seed pulse was amplified more than 3 times. As  $I_2$  approached  $I_1$  the ratio dropped to  $\sim 1.$ 

For the 532 nm pumping pulse focused to  $10^{13}$  W/cm<sup>2</sup>, the vector potential is  $a_1 \approx 0.0014$ , therefore the growth rate, calculated by Eq. (2), is  $\gamma_{RBS} \approx 1.3 \times 10^{12} \text{ s}^{-1}$ . If there is no energy loss and the amplification occurs through the whole Rayleigh length of the pumping beam ( $Z_R \approx 0.6$  mm), for a low intensity seed pulse (no pump depletion – linear regime) one would expect an amplification of  $exp(2\gamma_{RBS}Z_R/c) \approx 180$ . However, given an estimated preplasma temperature  $T_e \approx 20$  eV and  $n_e = 3 \times 10^{20}$  cm<sup>-3</sup>, the inverse Bremsstrahlung absorption length is ~100 µm, i.e. the penetration distance of the pumping pulse into the plasma is only 100-200 µm. The actual situation is more complicated since the plasma is heated and the plasma density is changed by the pumping pulse. In later density measurements it was found that the plasma does not fill the whole microcapillary at short delays, hence the microcapillary length is not the

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actual interaction length. If the effective interaction length is  $L_{eff} \sim 200 \ \mu m$ , the amplification would be  $\exp(2\gamma_{RBS}L_{eff}/c) \sim 6$ , which would be in good agreement with the observed amplification.

The short effective interaction length can also explain the decrease of the amplification with increasing seed pulse intensity. During the measurement of Raman amplification as a function of seed pulse intensity (at constant pumping intensity  $I_1 \approx 10^{13}$  W/cm<sup>2</sup>) the amplification dropped as the intensity of the seed pulse (I<sub>2</sub>) approached that of the pumping pulse (I<sub>1</sub>). The explanation in view of the short L<sub>eff</sub> could be as follows. The available pumping energy within the short interaction time (for L<sub>eff</sub> = 0.2 mm the interaction time is  $\tau_{eff} = 2L_{eff}/c \approx 1.5$  ps) of the total 150 mJ in 5 ns is only ~ 40  $\mu$ J. When I<sub>2</sub>  $\approx$  I<sub>1</sub>, the energy of the seed pulse is ~ 30  $\mu$ J, which is comparable to the available pumping energy. Even with total pump depletion one cannot expect the amplification to exceed a factor of 2. While taking into account absorption and no full depletion, there should be no visible amplification.

#### Experiment with a broadband Seed Pulse in Copper Microcapillaries

In order to check our hypothesis that in the 3 mm long microcapillary, only  $\sim 200 \,\mu$ m of the plasma had the matched high density, we continued the experiment with much shorter microcapillaries [10]. This allowed us to have a more precise knowledge of the interaction length and to avoid any refractive effects.

The experimental setup was the same as in Fig. 1, except that the seed pulse was provided by an optical parametric oscillator (OPO), which was pumped by the 745 nm subpicosecond pulse and produced a very broadband output pulse in the visible region. Since the broadband seed pulse has a spectrum with some random peaks and the spectral distribution varies from shot to shot, a small portion of the seed pulse was split out and focused into the spectrometer to provide a reference spectrum. Fig. 3 (a) displays the reference spectrum (top) and the seed pulse spectrum after passing through the empty microcapillary (bottom), demonstrating a good correspondence to each other. In order to minimize the spatial mismatch of the seed and the pumping pulses, as encountered in the earlier experiment, the microcapillary was imaged at the entrance slit of the spectrometer. The related geometry is shown in Fig. 3 (c). The pumping pulse was focused onto the center of the microcapillary with  $\sim 20 \ \mu m$ FWHM ( $Z_R \sim 0.6 \text{ mm} > L_{cap}$ ). The seed pulse was slightly defocused to make sure that part of the seed overlaps with the pump. The entrance slit was closed to a width of 200  $\mu$ m, corresponding to 30  $\mu$ m in the microcapillary plane, to cover the interaction area between the seed and the pump pulses. With such an arrangement, the spectrum displayed by the CCD camera had a spectral resolution of  $\sim 0.6$  nm and a spatial resolution of  $\sim 10 \ \mu\text{m}$  in the vertical direction [marked as the y-axis in Fig. 3 (b) and (c)].

The prepulse for creating the preplasma inside the microcapillary and the pumping pulse were the same as in the previous experiment, i.e. about 60 mJ at 248 nm in 20 ns and 150 mJ at 532 nm in 5 ns, respectively. The spectrally resolved amplification was calculated as  $I_p(\lambda) / I_0(\lambda)$ , where  $I_p(\lambda)$  is the intensity of the amplified pulse spectrum and  $I_0(\lambda)$  is the intensity of the reference spectrum. Without the plasma the ratio  $I_p / I_0$ 

is approximately a flat curve. When the plasma is created in the microcapillary, the seed pulse spectrum remains almost the same, with small spatial modifications, probably due to nonuniformities of the plasma density. When the pumping pulse is fired into the plasma, the seed pulse spectrum demonstrates a significant difference between the center,  $I_p$  ( $\lambda$ , y = 0), and the edge,  $I_p$  ( $\lambda$ ,  $y = 100 \,\mu$ m), as shown in Fig. 3 (d) (for a copper microcapillary with  $L_{cap} = 200 \,\mu$ m,  $D_{cap} = 150 \,\mu$ m and the delay between the prepulse and the pumping pulse was 40 ns). The ratio  $I_p$  / $I_0$  at the central part has a distinguished peak at  $\lambda = 645-650$  nm with a maximum of ~ 7, while the ratio at the edge of the plasma channel is much flatter and does not exceed 2 for most wavelengths. The observed maximum amplification is 3-4 for copper microcapillaries with  $L_{cap} = 200 \,\mu$ m and  $D_{cap} = 150 \,\mu$ m. The intensity of the seed pulse was below 10<sup>11</sup> W/cm<sup>2</sup> thus there was no significant pump depletion, i.e., we were in the linear regime for this experiment. The linear growth rate of Raman backscattering is calculated to be  $\gamma_{RBS} \approx 1.0 \times 10^{12} \, \text{s}^{-1}$  for  $I_{pump} = 1 \times 10^{13} \, \text{W/cm}^2$  and  $n_e = 1.3 \times 10^{20} \, \text{cm}^{-3}$  (the density corresponding to 645 nm). The amplification predicted by the linear theory for  $L_{cap} = 200 \,\mu$ m is then exp ( $2L_{cap} \,\gamma_{RBS}/c$ )  $\approx 3.8$ , which is in good agreement with the experimental observation.



**FIGURE 3.** (a) Reference spectrum (top,  $I_0$ ) and the seed pulse spectrum (bottom,  $I_p$ ). (b) Reference spectrum and the amplified pulse spectrum in a copper microcapillary with  $L_{cap} = 200 \ \mu m$  and  $D_{cap} = 150 \ \mu m$  at a delay of 40 ns. (c) Geometry relation at the entrance slit of the spectrometer. (d) Ratio of  $I_p$  ( $\lambda$ )/ $I_0$  ( $\lambda$ ) at the center area (y = 0) and at the edge (y = 100  $\mu m$ ), both integrated over  $\Delta y = 30 \ \mu m$ , for the spectra shown in (b).

#### CONCLUSION

In conclusion, we have shown Raman amplification of ultrashort laser pulses by a factor of ~ 6 in microcapillary plasmas. The analysis of the experimental data in LiF microcapillaries, together with simulation results [10], indicates that the length of the plasma with the matched density,  $n_e = 3 \times 10^{20} \text{ cm}^{-3}$ , was much shorter than the microcapillary. We conclude that due to this short effective length the observed amplification in our initial experiment was significantly less than the linear theory predicted, and the amplification dropped as the energy of the seed pulse increased since the available pumping energy within this narrow time window was very limited. Further investigations in 200 µm-long microcapillaries support the conclusion of short interaction lengths and the results show an agreement between the observed amplification and the linear growth rate.

In order to obtain larger amplification a longer plasma column with proper density is desired. Longer copper microcapillaries ( $L_{cap} = 500 \ \mu m$ ) were found unsuitable since the plasma could not fill the whole microcapillary, due to the illumination geometry of the prepulse. An alternative method for creating longer plasma is a high-pressure gas jet, which allows for 2-D density measurements since it is an open system. The experiment of Raman amplification in a gas jet plasma has confirmed Raman resonance by simultaneous density measurements.

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