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Stimulated Raman scattering of XeCl 70 ns laser pulses in silica fibres

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Abstract. For the first time stimulated Raman scattering (SRS) in multimode silica fibres pumped with relatively long XeCl laser pulses (70 ns) has been observed. The spectral and temporal development of SRS has been studied both experimentally and theoretically. Two orders of SRS have been obtained in the 311–317 nm spectral region. The experimental results are in good agreement with computer simulations of SRS in which the effects of self- and cross-phase modulation and group velocity dispersion are negligible. Included in the numerical model are experimentally measured spectra of the Raman gain curve and the pump laser spectra.

Keywords: Raman scattering, excimer laser, optical fibre

1. Introduction

Many of the applications of excimer lasers require their radiation to be delivered through optical fibres [1]. For the transmission of high-energy pulses, optical fibres with low losses in the UV region and excimer lasers generating relatively long optical pulses have been developed. The literature does not give sufficient data for the nonlinear optical effects in optical fibres related to the propagation through them of long XeCl laser pulses. This reflects directly in practical solutions where high-energy pulses are delivered through optical fibres.

On the other hand, the optical fibres can be used successfully as frequency shifters and amplifiers, not only as passive transmission lines. Fibre Raman lasers in the UV region are attractive as efficient solid-state frequency converters [2].

In this paper we have examined the spectral and temporal characteristics of Stokes pulses pumped by 70 ns XeCl excimer laser pulses in multimode optical fibres. All fibres that we have used (CeramOptec, Fiberguide, PolyMicro Technology) are OH^- rich and are suitable for transmission in the UV region.

A computer simulation analysis based on coupled differential equations was used to represent stimulated and spontaneous Raman scattering. The simulations, confirmed by experiment, show the evolution of the first and second Stokes spectra and their pulse forms.

2. Experiment

2.1. Experimental setup and apparatus

The excimer laser used to pump the fibres is a laboratory-made XeCl laser [3]. The excitation scheme provides direct storage capacitor drive, high-voltage discharge prepulsing and automatic UV preionization. The laser chamber was filled with a gas mixture of HCl:Xe:Ne = 3:30:3000 mbar. The optical cavity was composed of two flat mirrors with reflectances of 100% and 40%. The device produced an output energy of over a 200 mJ in ≈65–70 ns optical pulses (FWHM).

The output spectrum of the laser was composed of two strong lines ($\lambda_1 = 307.96$ nm and $\lambda_2 = 308.21$ nm) and two weak lines ($\lambda_3 = 307.70$ nm and $\lambda_4 = 308.46$ nm). The energy distribution measured in our spectral studies was approximately $\lambda_1{:}\lambda_2{:}\lambda_3{:}\lambda_4 = 43{:}55{:}1.8{:}0.2$.

The optical fibres used in the experiments were step-index, multimode, OH^--rich silica fibres, suitable for transmission in the UV spectral region (CeramOptec, Fiberguide and PolyMicro Technology), with a core diameter of 200 μm and a cladding diameter of 220 μm, a numerical aperture (NA) of 0.2 and a fibre length of 70 m.

The output laser beam was focused onto the fibre with a lens of 50 mm focal length. The input face of the optical fibre was placed 1–2 mm behind the focus to avoid optical damage. The output light was collimated with a 50 mm focal-length lens, dispersed with a 3600 lines mm^{-1} diffraction grating, detected by fast PIN photodiodes and stored by a two-channel oscilloscope—Tektronix-466 (100 MHz).

The output energy of the excimer laser was measured with a Gen-Tec ED-100 joulemeter. We measured the input

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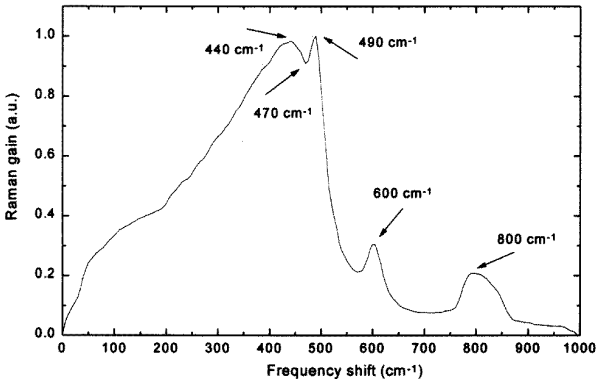

Figure 1. Raman spectrum of Fiberguide SFS 200/220T silica fibre.

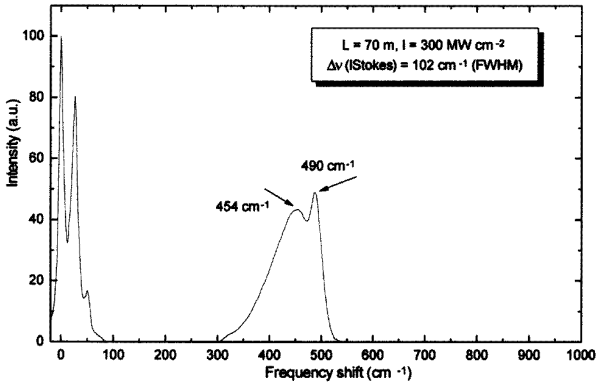

Figure 2. The output spectrum from a 70 m long optical fibre pumped with $I = 300$ MW cm^{-2} of XeCl laser pulse.

energy in the fibre and the threshold energy of the stimulated Raman scattering (SRS) with the cutback method. The spectrum of the fibre output was analysed with a prism spectrograph (ISP-30) with a 1.6 nm mm^{-1} dispersion and an input slit of 20 μm. The registration was performed on a photoplate ORWO-WU-3 in a single-shot regime. We measured the spontaneous Raman spectra for all three types of optical fibres. The spectra were obtained using 488 nm excitation with a SPEX 1404 that employed a cooled RCA C31034 photomultiplier tube. For the frequency interval of 1000 cm^{-1} all the spectral profiles were identical and figure 1 shows one example. We used this profile in our calculations described in section 3. The characteristic features of the gain curve are the broad peak around 440 cm^{-1} and the higher and sharper one at 490 cm^{-1}. There are also two low peaks at 600 and 800 cm^{-1}.

2.2. Experimental results

We studied the process of SRS for all three types of optical fibres. The obtained results were very similar in their Stokes spectral profiles which we explained as being due to the similar composition of the fibres. The only difference was in the threshold of SRS process, which was due to the different linear losses.

The experimental results shown in this section were obtained with Fiberguide optical fibre. The low-intensity attenuation of this fibre measured by the cut-back method was 160 dB km^{-1} at 308 nm. We determined the thresholds of the first and the second Stokes generation as 200 and 400 MW cm^{-2}, respectively. The damage threshold was 650 MW cm^{-2} which was close to the threshold of the third Stokes.

In figures 2 and 3 we present the output spectra for three different pump intensities. The spectrum of the first-order Stokes (figure 2) has a two-peak structure with a form that corresponds very closely to that of spontaneous Raman scattering. The Stokes waveforms for higher input intensities are shown in figures 3(*a*) and (*b*).

We observed (1) narrowing of the spectral width (FWHM) with increase of pump power; (2) red-shift of the first peak (440 cm^{-1}) of the first Stokes but no similar shift for the second peak (490 cm^{-1}); (3) the same shift effect for the peak of the second Stokes which has only a one-peak spectral profile.

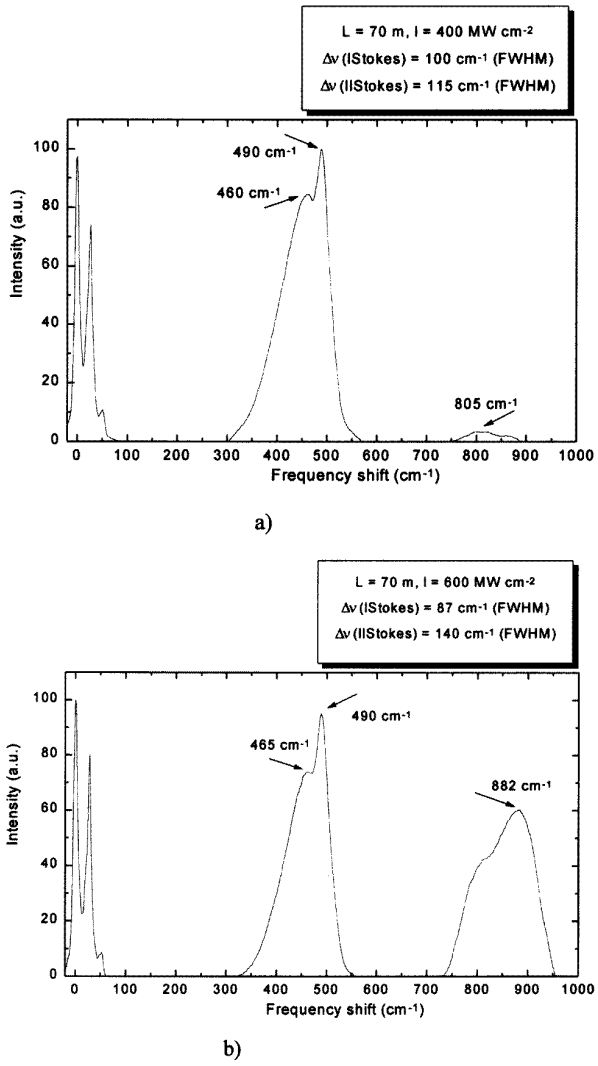

Figure 3. The output spectra from a 70 m long optical fibre pumped with two different XeCl laser input intensities: (*a*) 400 MW cm^{-2} and (*b*) 600 MW cm^{-2}.

The red-shift effect is due to the energy transfer from the shorter wavelengths to the longer under the Raman gain profile. This effect was not observed for the second peak of the first Stokes because of the strong decrease of the

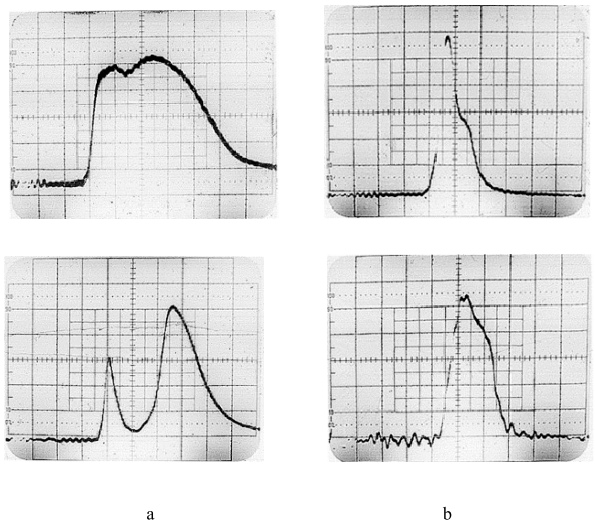

Figure 4. The remaining pump pulse (*a*) and the first Stokes pulse (*b*) at the end of the fibre (scale 20 ns/division).

Raman gain after 490 cm^{-1}. The disappearance of the two-peak structure in the form of the second Stokes is due to the convolution effect between the broad first Stokes and the spectrum of the Raman gain. In figure 3(*a*) the spectrum of the SRS near the threshold of the second Stokes generation is presented. Its maximum was at 805 cm^{-1} and with increased pump power (figure 3(*b*)) this maximum shifted to 882 cm^{-1}. At low intensities the second Stokes started to grow at 800 cm^{-1}. This corresponds very closely to the weak peak in the Raman gain profile. With increase of the pump power the role of the first Stokes as a pump for the second becomes essential.

It is useful to mention that in all our spectra we observed a two-peak structure of the first Stokes. We explain this specific profile as being due to the convolution of the XeCl multi-line output spectra and the Raman gain profile. For example, the position of the first peak has been determined from the two strong lines of the pump spectra since the position of the second depends mainly on the Raman gain profile and the second strong line of the pump. This effect does not exist in the experimental results of other groups in this field of investigation [2, 7]. We investigate these phenomena theoretically and the results are shown in section 3 of this paper.

Next, we observed the waveforms of the pump and the first Stokes by pin photodiodes and a storage oscilloscope, with rise times of 1 and 3 ns, respectively. The results are shown in figure 4.

The waveforms of the depleted pump pulse are wider compared with the input pulse of 7–8 ns (figure 4(*a*)). This is mainly due to modal dispersion of the fibre. The maximum modal dispersion for multimode fibres can be estimated by the difference between the transmission time of pulses along the core and the cladding—about 5 ns for our 70 m long sample of optical fibre. We roughly estimated this modal-dispersion time along the core and the cladding, when the incident angle of the light on the fibre was distributed within the full NA.

Figures 4(*a*) and (*b*) show the weak and strong depletion of pump intensities (left column) and corresponding Stokes pulses (right column).

The waveform of the remaining pump has two peaks because the Stokes starts to increase in the maximum of the pump. In the regime of strong SRS the middle of the pump is fully depleted and the pump at the end of the optical fibre essentially consists of two separated pulses, very different in intensity and duration. We observed that the first peak was always weaker and narrower than the second peak. We consider that the reasons for this observation are as follows: the group velocity dispersion of the Stokes and pump pulse because of the modal and wavelength dispersion. Although this dispersion is small compared with the pulse duration the leading front of the pump pulse grows quickly and therefore the depletion is stronger.

The duration of the first Stokes pulses expanded with the increase of the input pump intensity—from 10–15 ns by the threshold of the first Stokes generation to 30–35 ns by the threshold of the second Stokes. The time behaviour of the second Stokes was similar.

Despite the results presented in the paper of M Perrone [7], where shortening of the total output laser pulse duration at the end of the fibre in the regime of the SRS was observed, we only obtained a pulse shortening for the Stokes pulses and not for the total output pulse. Naturally, the shortest Stokes pulses we observed were at the threshold of the SRS process.

3. Theoretical analysis

To examine the evolution of the first and second Stokes spectra we have carried out an analysis by solving coupled differential equations. We assumed that for our pulse duration ($\approx$70 ns FWHM) and fibre length ($L = 70$ m) the pulse walk-off, the group velocity dispersion and self- and cross-phase modulation were negligible. The pulse excitation was carried out as a superposition of a series of CW calculations at different input intensities. We approximate the pump pulse with equally long slices of 1 ns with constant intensity. For CW calculations we divided the spectral interval in Raman gain into discrete frequencies of 5 cm^{-1}.

Our numerical calculations were based on laser and fibre characteristics used in the experiment described above.

We used a moving coordinate system that travels with the group velocity of the pulse. In the system, I_i is the intensity at the ith frequency interval; α_i and α_{NL} are the linear and nonlinear absorption at the corresponding frequencies ($\alpha \sim \lambda^{-4}$); g_{jk} is the Raman gain at k, pumped by frequency j; g_m is the maximum of the Raman gain and $C_i = R\pi(\mathrm{NA})^2(\beta/\lambda_i^4)$ is the spontaneous Raman scattering, where $R\pi(\mathrm{NA})^2$ is the fraction of the light scattered into 1 sr captured by the guide and $\beta = 1.5 \times 10^{-27}$ cm^4 is the differential cross section of the spontaneous Raman scattering. For multimode optical fibres all the light emitted within a solid angle given by the fibre's NA is guided so the factor $R = 1$ [4]. We included the nonlinear absorption in the equations because for our input intensities it became comparable with the linear absorption [5]:

$$\frac{\mathrm{d}I_0}{\mathrm{d}z} = -(\alpha_0 + \alpha_{\mathrm{NL}} I_0) I_0 - I_0 \sum_{k=1}^{n} g_{0k} I_k - \sum_{k=1}^{n} C_0 I_0 \frac{g_{0k}}{g_m} \Delta\nu$$

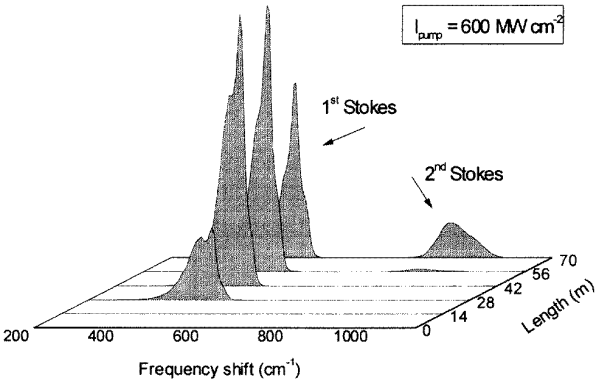

Figure 5. Calculated evolution of the Stokes spectra ($I_{\text{pump}} = 600$ MW cm^{-2}).

$$\frac{\mathrm{d}I_i}{\mathrm{d}z} = -(\alpha_i + \alpha_{\text{NL}} I_i) I_i + I_i \sum_{j=0}^{i-1} g_{ji} I_j - I_i \sum_{k=i+1}^{n} g_{ik} I_k + \sum_{j=0}^{i-1} C_j I_j \frac{g_{ji}}{g_m} \Delta\nu$$

$$\frac{\mathrm{d}I_n}{\mathrm{d}z} = -(\alpha_n + \alpha_{NL} I_n) I_n + I_n \sum_{j=0}^{n-1} g_{jn} I_j + \sum_{j=0}^{n-1} C_j I_j \frac{g_{jn}}{g_m} \Delta\nu.$$

The pump pulse spectrum that we used was very close to the real spectrum of the laser. In our model the pump laser spectrum was presented with three wavelengths: $\lambda_1 = 307.96$ nm, $\lambda_2 = 308.21$ nm and $\lambda_3 = 308.46$. The energy distribution between these lines was as follows: $\lambda_1{:}\lambda_2{:}\lambda_3 = 55{:}43{:}2\%$. The spectral width (FWHM) that we used was the same as the measured value.

We measured the linear absorption for λ_1 which for our fibre (Fiberguide) was $\alpha = 3.5 \times 10^{-4}$ cm^{-1}. For the nonlinear absorption coefficient we used the value $\alpha_{\text{NL}} = 4.6 \times 10^{-7}$ cm MW^{-1} for the whole spectral region of the pump and Raman scattering [5].

In this spectral region (308 nm) the reports for Raman gain g_m were very different and we made some calculations with different Raman gain coefficients to ensure good agreement with our experiments. The obtained value of the gain was $g_m = 3 \times 10^{-13}$ m W^{-1}, very close to the value calculated by the relation $g \sim \lambda^{-1}$ taken from Stolen *et al* [6].

For demonstrative purposes, in figure 5 we show the sequence of Stokes generation along the fibre length. In this case we used a CW calculation with an input pump intensity of 600 MW cm^{-2}. This figure presents the evolution of the Raman shape for two generated Stokes—red-shift of the peaks and narrowing of the spectral width.

Figure 6 are shows typical spectra of the first and second Stokes in the regime of SRS. Three kinds of SRS spectra are presented: (*a*) the SRS spectrum when the pump power is near to the threshold of the first Stokes generation; (*b*) when we have a process of significant conversion of the pump to the first Stokes and (*c*) when the pump power is enough for the second Stokes generation.

We note that there is good agreement with the experimental spectra: (1) the position of the first peak is shifted from 440 cm^{-1}; (2) the peak at 490 cm^{-1} is not shifted

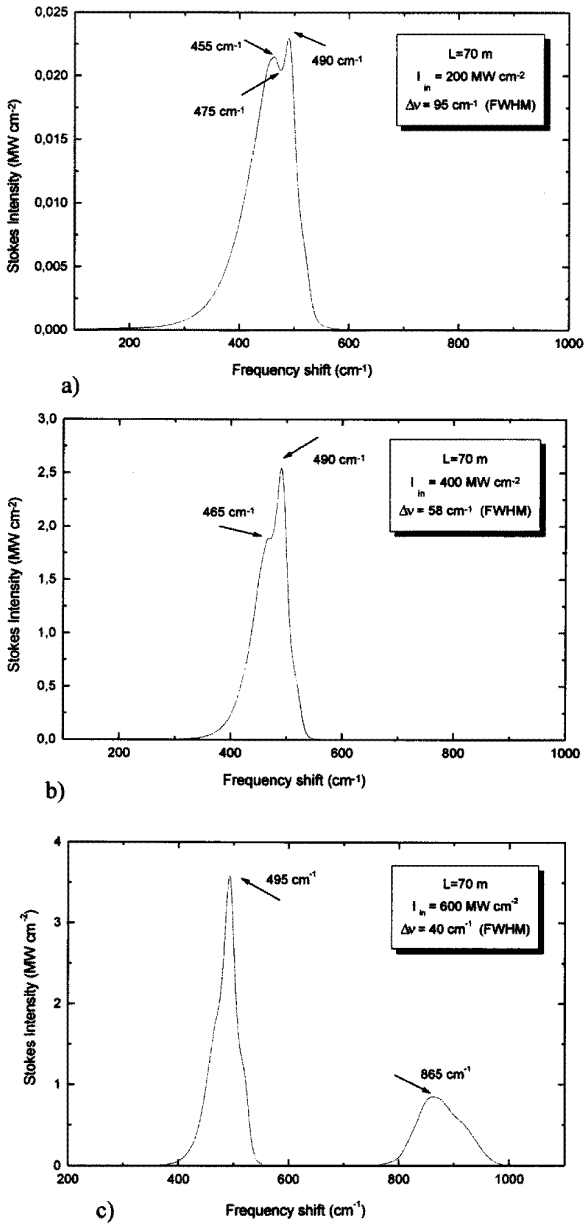

Figure 6. The calculated spectra of SRS for three different values of the pump intensity: (*a*) 200 MW cm^{-2}, (*b*) 400 MW cm^{-2} and (*c*) 600 MW cm^{-2}.

irrespective of the pump power. With increase of the pump power our model shows 'red-shift' of the first peak in the shape of the first Stokes and this finally lead to a form with only one peak at 490 cm^{-1}. Our calculations confirmed that the form of the second Stokes is more symmetric than the first Stokes shape.

Our model also gives information about the time waveforms of the pump and Stokes pulses. In the calculation we used the real shape of the laser input pulse. An example of the results of our calculation is shown in figure 7. The results are plotted as the total energy of the output pulse versus frequency shift (by integrating over time the intensity for each frequency interval of the calculation).

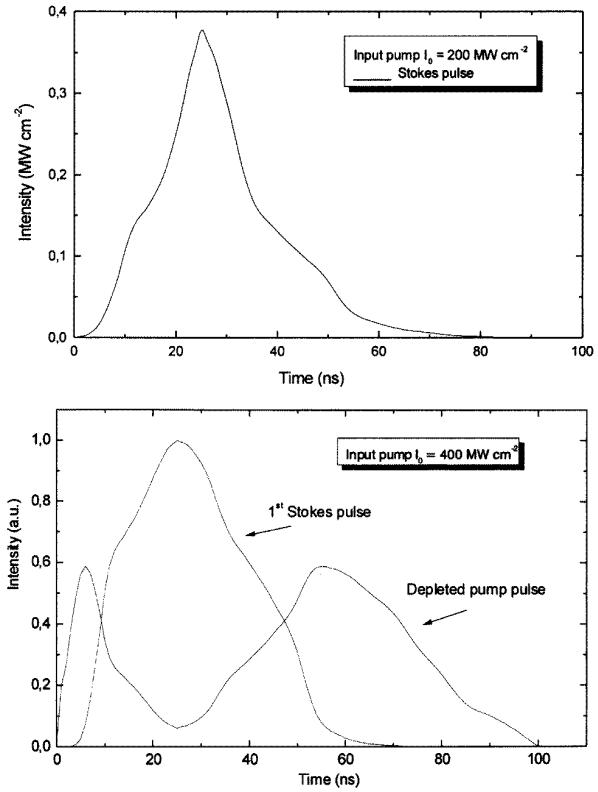

Figure 7. The pulse forms of the Stokes and depleted pump pulse.

4. Conclusion

We have obtained two orders of Stokes pulses of SRS in optical fibres pumped by relatively long (70 ns) XeCl laser pulses. The input intensity dependence was studied for both the spectral and time behaviour of the first and second Stokes. The computer simulation reproduced these experimental results well. They coincide in (1) the red-shift for the first and second Stokes; (2) the asymmetric shape and symmetric shape of first and the second Stokes spectral forms; (3) the time waveforms of the depleted pump and Stokes pulses generated. In summary, we hope that this work will be useful for estimating the influence of SRS on the capability of fibre optic delivery in the UV spectral region.

Acknowledgment

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