Broadband-tunable LP₀₁ mode frequency shifting by Raman coherence waves in H₂-filled hollow-core PCF: supplementary material

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Published 1 June 2015

This document provides supplementary information to “Broadband-tunable LP₀₁ mode frequency shifting by Raman coherence waves in H₂-filled hollow-core PCF,” <http://dx.doi.org/10.1364/optica.2.000536>. We describe the theoretical model employed to simulate the dynamics of the system and show the individual photon rates of the first two up-shifted sidebands (M_1 and M_2), where only their sum was shown in the main text. In addition, we also provide details on the pressure dependence of the interference of two coherence waves. © 2015 Optical Society of America

<http://dx.doi.org/10.1364/optica.2.000536.s001>

1. Numerical simulations

To simulate the spatio-temporal dynamics of the C_w amplitude Q and the optical fields, we used a semi-classical treatment of stimulated Raman scattering [1] based on the following set of coupled Maxwell-Bloch equations:

$$\frac{\partial}{\partial z} E_l = -i\kappa_{2,l-1} Q E_{l-1} q_{l-1} q_l^* \frac{\omega_l}{\omega_{l-1}} - i\kappa_{2,l} Q^* E_{l+1} q_{l+1} q_l^* - \frac{1}{2} \alpha_l E_l \quad (\text{S1})$$

$$\frac{\partial}{\partial t} Q = -Q/T_2 + in \frac{1}{4} \sum_l \kappa_{1,l} E_l^* E_{l+1} q_{l+1}^* q_l, \quad (\text{S2})$$

where the integer index l denotes the Raman sideband of the pump at frequency $\omega_l = \omega_w + l\Omega_R$ (ω_w is the frequency of the pump). The complex electric field amplitudes $e_l(z, \tau)$ are described in terms of the slowly varying envelopes $E_l(z, \tau)$:

$$e_l(z, \tau) = E_l(z, \tau) q_l = E_l(z, \tau) e^{i\beta_l z} \quad (\text{S3})$$

where the propagation constants β_l are calculated using Eq. 2 in the main text. The parameters α_l are the attenuation constants of the different spectral lines. We assume the population difference is $n = -1$, i.e., the majority of the molecules remain in the ground state, which is a reasonable approximation since the highest value of coherence $|Q|$ reached in the experiments is ~ 0.01 . The

frequency-dependent coupling coefficients $\kappa_{1,l}$ and $\kappa_{2,l}$ can be derived from the experimentally determined gain values γ [2] by:

$$\kappa_{1,l} = \sqrt{\frac{2\gamma c^2 \epsilon_0^2}{NT_2 \hbar \omega_l}} \quad (\text{S4})$$

and

$$\kappa_{2,l} = \frac{N \hbar \omega_l \kappa_{1,l}}{2\epsilon_0 c}, \quad (\text{S5})$$

where N is the molecular number density, c is the speed of light in vacuum, ϵ_0 is the vacuum permittivity, $\hbar$ is Planck's constant and T_2 is the dephasing time of the molecules, given in terms of the linewidth $\Delta\nu \sim 1/T_2\pi$ of the Raman transition [2, 3].

To model the dynamics of the mixing beam fields E_m ($\omega_m = \omega_M + m\Omega_R$), where m is an integer) we used the same set of coupled field equations as for the pump (Eq. S1), except that Q takes the value set by SRS in the preparation stage. Since the mixing fields are weak, they are assumed to have no effect on the C_w amplitude (for stronger mixing fields significant amplification or attenuation of Q will occur). Also, since the pulse lengths are comparable to the fiber length (1 m) group velocity walk-off between the different spectral lines is negligible.

The first Stokes signal is generated from noise in the experiment, initiating all the observed dynamics. In the simulations we assume a uniform noise floor of $E_{\text{noise}} = 500$ V/m for all the Raman lines except $l = 0$. We checked the validity of this

assumption by measuring the Stokes intensity as a function of pump intensity and extrapolating the exponential growth of the Stokes signal for low pump intensities. This yielded a value close to $E_{\text{noise}} = 500 \text{ V/m}$ for zero pump signal.

The initial shape of the W_1 pulse was assumed to be Gaussian and the modal intensity distribution was modeled by a uniform transverse profile with an effective mode area of $A_{\text{eff}} = 1.5a^2$ [4], where a is the core radius in the kagomé-PCF. Finally, we considered the same pump energies in both simulations and experiments.

The attenuation coefficients at the wavelengths used in the experiments were extracted from the loss measurement shown in Fig. S1.

Fig. S1. Measured loss of the HC-PCF (solid line) with estimated error (shaded region). Wavelength ranges without data have loss $> 5 \text{ dB/m}$ or no loss data are available. The frequency bands used in the Letter are highlighted by the vertical dashed lines.

2. Separate photon rates of up-shifted sidebands

Fig. S2. Experimental and theoretical normalized photon rates of the first (M_1) and second (M_2) mixing sidebands, plotted against increasing pressure (normalized to the maximum of the M_0 rate, not shown) for pump pulse energies E_p of 20 and 30 μJ . Also the sum of both signals ($M_1 + M_2$) is included which was shown in Fig. 3.

In the Letter we restrict the discussion on the dynamics of the mixing beam to the $M_0 \rightarrow M_1$ conversion process, although subsequent scattering to the second sideband ($M_1 \rightarrow M_2$) is also possible (see also Fig. 7 in the main text). Therefore we plot in Fig. 4 of the main text only the sum of the two upshifted signals ($M_1 + M_2$) for clarity. In Fig. S2 we show for completeness the photon rates of the two individual signals together with their sum. Due to the relatively flat dispersion at the phase-matching pressure of the $M_0 \rightarrow M_1$ transition (12 bar, see Fig. 2 in the main text) photons can subsequently be up-shifted from the first to the second sideband giving rise to the M_2 signal in Fig. S2 at $\sim 12 \text{ bar}$. In contrast, at 30 bar the dispersion is no longer flat and scattering to the M_2 band is strongly suppressed. No significant M_3 signal was observed in our experiments.

3. Coherence wave interference

The periodic pattern along z observed in the coherence at 27 bar in Fig. 5 is caused by interference of the two coherence waves excited in the system. The observed beat length $L_B = 3.9 \text{ cm}$ agrees well with the value predicted from the dispersion relation for the light, given by:

$$L_B = \frac{2\pi}{\Delta\beta_{(W_0 \rightarrow W_{-1})} - \Delta\beta_{(W_{-1} \rightarrow W_{-2})}}. \quad (\text{S6})$$

Fig. S3 shows how these interference fringes change with pressure and propagation distance for a pump pulse energy of 30 μJ and $\tau = 2 \text{ ns}$. At the dual phase-matching pressure of 12 bar, $L_R \rightarrow \infty$ and no fringes are visible along z , whereas for increasing pressures spatial oscillations of $|Q|$ are observed. These oscillations decrease in period as the pressure (and therefore the phase-mismatch) increases. In addition, the onset of the $W_{-1} \rightarrow W_{-2}$ coherence wave is also affected by the increasing phase mismatch; as the pressure increases, this C_w moves closer to the fiber endface because coupling from W_{-1} to W_{-2} by the $W_0 \rightarrow W_{-1}$ coherence wave becomes less efficient. Thus, the phase-matched conversion process at 12 bar turns smoothly into a cascaded process where the second Stokes band is generated via SRS from the first Stokes band. Note that the growth of the C_w amplitude is retarded for pressures below 10 bar because of decreasing gain, which prevents clear identification of the interference fringes.

Fig. S3. Amplitude of the coherence $|Q|$ at fixed time $\tau = 2 \text{ ns}$ as a function of pressure and propagation distance. The dashed vertical line marks the pressure where the two C_w 's are identical.

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