Resonant dispersive wave emission in hollow capillary fibres filled with pressure gradients

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Resonant dispersive wave (RDW) emission in gas-filled hollow waveguides is a powerful technique for the generation of bright few-femtosecond laser pulses from the vacuum ultraviolet to the near infrared. Here we investigate deep-ultraviolet RDW emission in a hollow capillary fibre filled with a longitudinal gas pressure gradient. We obtain broadly similar emission to the constant-pressure case by applying a surprisingly simple scaling rule for the gas pressure and study the energy-dependent dispersive-wave spectrum in detail using simulations. We further find that in addition to enabling dispersion-free delivery to experimental targets, a decreasing gradient also reduces the pulse stretching within the waveguide itself, and that transform-limited pulses with 3 fs duration can be generated by using short waveguides. Our results illuminate the fundamental dynamics underlying this frequency conversion technique and will aid in fully exploiting it for applications in ultrafast science and beyond.

Laser sources delivering bright, wavelength-tuneable few-femtosecond pulses are a key goal in ultrafast photonics with wide-ranging applications, for instance in time-resolved spectroscopy [1, 2]. Resonant dispersive wave (RDW) emission in gas-filled hollow-core waveguides is a particularly promising approach to achieving this goal, offering wide, continuous tunability from the vacuum ultraviolet [3, 4] to the near infrared [5] with high conversion efficiency and few-femtosecond duration. This technique is based on soliton self-compression, which occurs when anomalous group-velocity dispersion continuously compensates for the chirp induced by self-phase modulation during nonlinear propagation of a laser pulse. It was first demonstrated in small-core anti-resonant fibres at the microjoule pump energy scale [6]. Recently, we demonstrated that the use of large-core hollow capillary fibres (HCF) allows for significant energy scaling [4, 5, 7] while eliminating the tuneability gaps inherent to anti-resonant fibres [8, 9]. To make the potential offered by RDW-based sources a reality, a full understanding of the complex dynamics underlying the frequency conversion is required. Furthermore, several technical challenges, most importantly the delivery of compressed RDW-generated laser pulses to an experimental target, have to be overcome.

Here we study resonant dispersive wave emission in hollow capillary fibres under the influence of a longitudinal gas pressure gradient along the waveguide. Increasing pressure gradients, where the entrance of the HCF is under vacuum, are commonly used when compressing high-energy laser pulses by post-compression—spectral broadening in gas-filled HCF with subsequent phase compensation by dispersive optics [10]. The absence of gas at the entrance removes nonlinear effects which disturb the coupling to the waveguide and the increasing nonlinearity along the waveguide partially compensates for propagation losses [11]. For RDW emission, decreasing gradients are of particular interest since they allow for dispersion-free delivery of generated pulses into a vacuum system [12]. In our experiments, we find that it is surprisingly easy to obtain similar RDW emission in the cases of variable and constant pressure by applying a simple scaling rule, though with significant differences in the energy-dependent spectrum of the generated pulses, as previously observed in gas-filled anti-resonant fibres [8]. Through numerical simulations, we illuminate the mechanism behind this behaviour. Finally, we study the energy-dependent duration of the RDW pulse. We find that a decreasing pressure gradient reduces pulse stretching during propagation even before the HCF exit, and that widely tuneable transform-limited pulses with 3 fs duration can be generated by using short waveguides.

The principle of the experimental setup is shown in Fig. 1. It is identical to that detailed in ref. [4], except for the added option to create a pressure gradient. In brief: pulses centred at 800 nm with a duration of 30 fs full width at half maximum (FWHM) generated by a titanium-doped sapphire laser amplifier are compressed to 7.3 fs FWHM duration by spectral broadening in a helium-filled stretched HCF [13] and subsequent phase compensation by reflection off of dispersive mirrors and transmission through silica wedges. The profile of the compressed pulse, measured via second harmonic generation frequency-resolved optical gating (SHG-FROG), is shown Fig. 1(a). A combination of a motorised half-wave plate and a Brewster-angle silicon plate acts as a broadband variable attenuator. The compressed pulses are then coupled into a second stretched HCF with 250 µm core diameter and a length of 3 m. This HCF is sealed into a gas cell at each end, which can be independently evacuated or filled with gas. At the pressures used in our experiments, a single roughing pump is sufficient to keep the pressure in the evacuated cell below 1 mbar. The output from this second stage is analysed with a calibrated spectrometer.

When one end of the HCF is evacuated, the pressure distribution is given by

$$p_i(z) = p_{\text{max}} \sqrt{\frac{z}{L}}, \quad p_d(z) = p_{\text{max}} \sqrt{1 - \frac{z}{L}},$$

(1)

where $p_i(z)$ and $p_d(z)$ denote an increasing and decreasing pressure gradient, respectively, $z$ is the position along an HCF with total length $L$, and $p_{\text{max}}$ is the pressure on the high-pressure end [see Fig. 1(b)]. In conventional HCF-based post-compression systems, the dispersion of the waveguide is often very weak, with dispersion lengths many times longer than the HCF [4].

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With the nonlinearity dominating, similar spectral broadening can be obtained with constant and variable pressure by matching the integrated nonlinear phase shift (also known as the B-integral). For the increasing gradient in eq. [1] this is given by

\[ \phi_{nl} = P_0 \int_0^L \gamma(z) \, dz = P_0 \gamma_{\text{max}} \int_0^L \sqrt{\frac{2}{L}} \, dz = \frac{2}{3} P_0 L \gamma_{\text{max}}, \]  

where \( P_0 \) is the peak power of the initial pulse, \( \gamma(z) \) is the nonlinear coefficient—proportional to the nonlinear refractive index \( n_2(z) \) and hence to the pressure \( p(z) \)—and \( \gamma_{\text{max}} \) is its value at the high-pressure end. The factor of \( 2/3 \) is obtained for both decreasing and increasing gradients. In a conventional post-compression system, the fill pressure therefore simply needs to be increased by a factor of \( 3/2 \) as compared to the constant-pressure case, where \( \phi_{nl} = P_0 L \gamma_{\text{max}} \).

Figure 2 shows output spectra from the HCF as the input energy is changed for three different cases: a constant argon pressure of 83 mbar, an increasing gradient from vacuum to 125 mbar, and a decreasing gradient from 125 mbar to vacuum. The constant-pressure data shows typical behaviour observed in many previous studies of soliton self-compression and resonant dispersive wave emission in gas-filled hollow waveguides [4][8][14]: as the energy is increased, the spectrum broadens dramatically, until at some point a dispersive wave is generated at a much shorter wavelength than the driving pulse. In this case, the RDW first appears at an energy of around 40 µJ and at 250 nm wavelength.

The pressure for the gradients is chosen as a factor of \( 3/2 \) higher than the constant pressure, as it would be for a post-compression system. In contrast to simple spectral broadening, where the pulse shape does not change significantly during propagation, soliton self-compression is a strongly dynamical process. How the pulse evolves depends critically on the strength of both the nonlinearity and the dispersion, and the simple scaling rule accounts for neither the change in dispersion nor the evolution of the pulse during propagation. It is therefore reasonable to expect the dynamics of the self-compression to be significantly different from the constant-pressure case. Instead, our experimental data shows broadly similar behaviour for both decreasing and increasing gradients. As shown in Fig. 2(c) and (e), the RDW is first generated at nearly the same driving pulse energy, and furthermore its central wavelength is very close to that shown in Fig. 2(a). This close correspondence is unexpected, and we anticipate that it will be useful in designing vacuum-coupled frequency conversion systems based on RDW emission, since it allows the use of the simple relations that determine the required HCF and pulse parameters in the case of constant nonlinearity and dispersion [4][7].

Significant differences between the three cases do appear in the energy-dependent RDW spectrum. For constant pressure [Fig. 2(b)], the RDW spectrum initially shifts to shorter wavelengths as the energy is increased, but this blue-shift saturates at energies above \(~60 \) µJ. In an increasing gradient [Fig. 2(d)], the energy-dependent blue-shift is much stronger, covering nearly 50 nm between 40 µJ and 80 µJ and only saturating around \(~100 \) µJ. This could be a useful tool in applications where fast spectral tuneability over a relatively small
range is required, since the driving energy can be changed much more quickly than the gas pressure. Finally, for a decreasing gradient [Fig. 2(d)], the RDW spectrum initially shifts in the opposite direction, to longer wavelengths, before moving back at high energies. The overall shift is much reduced and comparable to the case of constant pressure. Similar behaviour has been observed in gas-filled anti-resonant fibre when using identical gas pressure for the three cases, leading to different RDW emission wavelengths [8]. Here, the similarities in the overall dynamics allow us to more directly compare the evolution of the dispersive wave.

To examine this behaviour more closely, we numerically simulate the RDW emission process. We use the field-resolved model detailed in ref. 4, which describes full multi-mode propagation in the HCF and includes the effects of dispersion and loss, the Kerr effect, as well as strong-field photoinionisation and plasma dynamics. There are no free parameters in this model—we use the measured input pulse shown in Fig. 1(a) and the coupled pulse energy as determined by the measured transmission of the evacuated HCF. The pressure gradient is described by eq. 1. The simulated output spectra for constant and decreasing pressure, shown in Fig. 3(a) and (b), faithfully reproduce the experimental data in Fig. 2(a-b) and (e-f), including the behaviour of the RDW spectrum.

As the driving pulse energy is increased, the stronger nonlinear interaction leads to faster self-compression. As shown in the left axis in Figs. 3(c) and (d), the self-compression length (SCL), defined here as the distance at which the self-compressing pulse reaches its maximum peak power, becomes much shorter. (Note that in the case of a decreasing gradient, the pulse always reaches its maximum peak power before the end of the HCF, since the near-zero nonlinearity near the exit prevents further self-compression.) The RDW is emitted very close to the self-compression point. In a gradient, this means that it is generated at different pressures depending on the energy, as shown on the right axis in Fig. 3(d). The combination of different pulse energy and different gas pressure then affects the phase-matching of the dispersive wave.

The wavelength of RDW emission is determined by the phase mismatch between the strongly nonlinear self-compressing soliton and a weaker linear (dispersive) wave. Their wavevectors can be approximated as

$$\beta_s(\omega) = \beta_0 + \beta_1 \Delta \omega + \frac{\gamma P_0}{2}$$

$$\beta_l(\omega) = \beta_0 + \beta_1 \Delta \omega + \frac{\beta_2}{2} \Delta \omega^2 + \frac{\beta_3}{6} \Delta \omega^3 + \cdots ,$$

where $\beta_s$ and $\beta_l$ are the soliton and linear wavevectors, respectively, and $\Delta \omega = \omega - \omega_0$ is the frequency detuning from the central frequency of the soliton $\omega_0$. The phase mismatch $\Delta \beta = \beta_1 - \beta_s$ for our experimental parameters is shown in Fig. 3(e). RDW emission occurs where the soliton and dispersive wave are phase-matched, i.e. $\Delta \beta = 0$. For 80 mbar of argon pressure (purple lines), the phase mismatch is zero at around 260 nm, close to the experimentally measured emission wavelength of 250 nm. At higher pressures (blue and green lines), the amount of anomalous dispersion is decreased and higher-order dispersion increased, leading to phase-matching at longer wavelengths. Higher peak power $P_0$ has the opposite effect [dashed lines in Fig. 3(e)], shifting the dephasing curve down by $\gamma P_0/2$ and leading to shorter phase-matching wavelengths.

The interaction between the changing self-compression length and the phase-matching explains the behaviour observed in our experiments. With constant pressure, the linear dispersion at the RDW emission point is also constant, and only the nonlinear shift acts on the spectrum. The resulting blue-shift, commonly observed in studies of RDW emission [6], slows at high energies since the peak power of the self-compressed pulse saturates: between 40 $\mu$J and 80 $\mu$J driving pulse energy, the maximum peak power more than doubles from 7.5 GW to 19 GW, but up to 120 $\mu$J it only increases to 24 GW. For an increasing gradient, faster self-compression means RDW emission at lower pressures. The nonlinear and linear contributions to the phase-matching thus add up, with both effects pushing the dispersive wave to shorter wavelengths. The resulting strong nonlinear dependence leads to the fast tuning observed in the experiment. In a decreasing gradient, the two effects compete instead, since RDW emission occurs at higher pressure for higher driving energy. Remarkably, these influences
The duration of the pulses in Fig. 4 is significantly longer than previously measured in anti-resonant fibres. Our simulations suggest that very broadband and nearly transform-limited pulses at a variety of wavelengths can be obtained by reducing the length of the HCF. Figure 5(a) shows output spectra as the energy is varied from a 1 m HCF with 250 μm core diameter filled with a decreasing pressure gradient. The spectral evolution is very similar to that observed in the 3 m HCF, though more energy is required to achieve RDW emission due to the shorter propagation length. Changing the gas pressure at the HCF entrance tunes the RDW wavelength.

In summary, we have shown that broadly similar resonant dispersive wave emission can be obtained in hollow capillary fibres filled with either constant gas pressure or a gradient. The very simple scaling rule of a factor 3/2 increase in gas pressure, adapted from pulse compression systems, is surprisingly successful in reproducing both the energy at which RDW emission first occurs and the central wavelength of the generated pulse. We have clearly identified the interplay between a change in self-compression length and the linear and nonlinear contributions to the dispersive wave phase-matching as the
mechanism behind the different spectral shifts of the dispersive wave that are observed for three pressure profiles. We have furthermore found that a decreasing pressure gradient is not only useful in eliminating the dispersion caused by transmission through windows, but also significantly reduces the pulse stretching within the waveguide itself. Finally, extremely short, transform-limited pulses can be generated by moving to shorter waveguides. We anticipate that our results will be very useful in exploiting RDW-based tuneable few-femtosecond light sources for cutting-edge time-resolved spectroscopy experiments as well as other applications.

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