

Supercontinuum generation in 1-decanol

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Abstract: Although solids have been recently used in ultrafast experiments for spectral broadening due to their relatively high nonlinearity, their sensitivity to damage limits their long-term stability. Liquids are a possible alternative to solids as a nonlinear medium because of their comparable nonlinearity and resistance to permanent damage. We generate a supercontinuum in 1-decanol that spans from approximately 450 nm to 950 nm. We measure the nonlinear index of refraction of 1-decanol and find a significant n_4 contribution. This contribution leads to a nonlinearity comparable to CS_2 (a frequent reference for nonlinear optics) in high-intensity regimes while being significantly less volatile and toxic. We find this supercontinuum spectrum to be stable for at least 30 minutes.

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1. Introduction

Ultrashort and few-cycle laser pulses require supercontinuum (SC) spectra spanning more than an octave of bandwidth [1]. Few-cycle laser pulses offer a route of coherent control on a femtosecond ($1 \text{ fs} = 1 \times 10^{-15} \text{ s}$) timescale, driving experiments in strong-field physics, high-harmonic generation, and attosecond science [2–6]. SC generation has been successfully achieved in gases [7–9] and in solids [10–12] with spectra spanning more than an octave. More recently, SC generation in liquids has been a growing area of study, particularly in hollow-core fibres [13] and liquid-core photonic-crystal fibres (PCFs) [14]. In bulk media, such broad spectra are often referred to as white light continua, as demonstrated for example by Manzoni et al. [15].

Liquids are favourable media for nonlinear optics due to their high density, high nonlinearity, and good power stability [16]. Ethanol has successfully generated SC in a PCF [17], although its volatility and low boiling point limit its stability [18]. Among the commonly used and effective liquids for SC generation are chloroform (CHCl_3) [19], carbon disulfide (CS_2) [20], toluene (C_7H_8) [14], and benzene (C_6H_6) [13]. A significant drawback of using these four liquids is that they are all highly toxic [21–24]. Due to the volatility and associated health risks with these organic solvents, we began studying high boiling point organic alcohols instead. The MAK ("maximale Arbeitsplatz-Konzentration" : maximum workplace concentration) [25] of 1-decanol is reportedly 10 mL/m^3 with no reported carcinogenicity [26], compared to a value of 5 mL/m^3 for carbon disulfide [27], 0.5 mL/m^3 for chloroform [28], 5 mL/m^3 for benzene [29], and 50 mL/m^3 for toluene [30] (but toluene vapours are known to be harmful in comparison to 1-decanol [31]).

We have previously studied the effects of 1-decanol on intense femtosecond pulses and found that its high boiling point (231.85°C) [32] leads to stable pulse compression by a factor of three that is readily achievable [33]. In this paper, we further explore the nonlinear optical properties of 1-decanol, demonstrating significant contributions from higher-order nonlinearities while generating white light continuum (WLC) spanning from 450 nm to 950 nm. We use the z-scan method to extract the Kerr nonlinearity in 1-decanol and find that it is significantly intensity dependent, which allows us to characterize the next order n_4 nonlinearity up to 10^{15}

W/m^2 . At higher intensities, we find that this higher-order contribution leads to a spatial optical soliton that dramatically increases the bandwidth by increasing the interaction region of high intensity without power loss. In addition, we demonstrate the stability of the WLC spectrum over 30 minutes, supporting long-term and repeated use of 1-decanol as a nonlinear medium for strong-field experiments.

2. Methods

As shown in Fig. 1(a), we use a Ti:Sapphire laser that produces 100 fs, 1.5 mJ pulses centered at 785 nm with 1 kHz repetition rate. The beam passes through a half-wave plate ($\lambda/2$) and a polarizer (Pol.) to control the power. A 200 mm focal length lens focuses the beam into the 1 cm cuvette of 1-decanol, which sits on an automated translation stage (Thorlabs PT1-Z8). The beam waist is measured to be 26 μm at the focus, giving a free-space Rayleigh range of $z_0 \approx 2.7 \text{ mm}$ and $z_R \approx 4 \text{ mm}$ inside the liquid [34]. When measuring the spectrum as a function of radial position, the spectrometer is placed 45 cm after the 200 cm focusing lens with no second lens after the cuvette so the beam is not focused into the slit of the spectrometer; instead, the spectrometer is scanned through the beam radially.

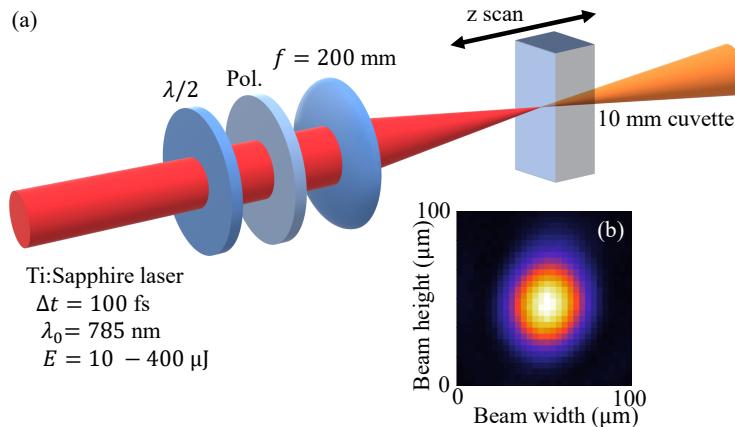


Fig. 1. (a) Experimental setup used for supercontinuum generation in 1-decanol. The beam travels from left to right passing through a half-wave plate ($\lambda/2$), followed by a polarizer (Pol.), a 200 mm focal length lens, and finally into a cuvette which sits on an automated translation stage. Not shown is the spectrometer that measures the spectrum of the output beam. (b) An image of the radial beam profile is shown.

In both the radial and axial cases, the data is taken using 3 wavelength filters and stitched together during data analysis to increase the dynamic range and to better resolve both the visible and infrared portions, which are relatively weaker in intensity compared to the central wavelength. To this end, the spectrum is measured with no filter in order to resolve the spectrum from 740-800 nm. Next, the same measurement is made using a visible filter (335-610 nm)(Thorlabs FGB37M), and two infrared (IR) filters (800-850 nm and 850 nm long-pass). Normalization is required to account for different spectrometer integration times and different degrees of beam attenuation before the spectrometer.

Filter transmission is corrected using measured transmission curves for each visible and IR filter. Because each filter isolates a defined passband, we retain only that wavelength region and stitch it to the unfiltered and visible spectra by matching their overlap in amplitude. A Savitzky–Golay filter is applied for mild smoothing before plotting. Our data and code are available in Ref. [35].

3. Results and discussion

3.1. Measuring the nonlinear index of refraction

We measure the nonlinear index of refraction of 1-decanol with the closed-aperture Z-scan technique [36]. The sample is translated through the focal region of the beam, and the on-axis transmittance through a small aperture is recorded as a function of position. The nonlinear phase shift accrued by the beam due to self focusing depends on the Kerr coefficient n_2 , the peak intensity I_0 , and the propagation distance within the material. Self-focusing modifies the beam divergence on either side of the focus, producing the characteristic curve shown in Fig. 2(a).

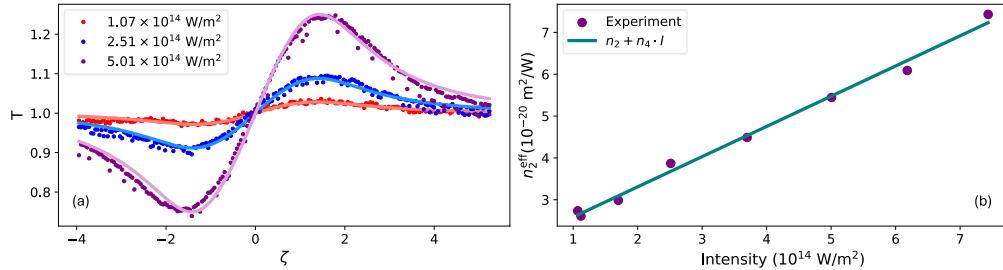


Fig. 2. (a) Low-intensity regime. The points are measured data while the solid lines are the corresponding curve fits. T is the transmittance, which is a function of normalized length $\zeta = z/z_0$ where z is the direction of beam propagation and z_0 is the Rayleigh range [33]. (b) Effective nonlinear index values $n_2(I_0)$ extracted from individual low-intensity Z-scan fits such as (a), plotted as a function of input intensity.

The close-aperture transmission is defined by the function,

$$T = 1 + \Delta\Phi F(\zeta, I) \quad (1)$$

where $\Delta\Phi = (2\pi/\lambda)n_2^{\text{eff}}z_0I_0$ is the self-focusing-induced nonlinear phase shift, λ is the laser central wavelength, n_2^{eff} is the effective nonlinear index coefficient [37], z_0 is the (free space) Rayleigh range, and I_0 is the peak intensity. The effective nonlinear index coefficient is defined by the third order (Kerr) nonlinear coefficient n_2 and the fifth-order coefficient n_4 , where $n_2^{\text{eff}} = n_2 + n_4I$. The characteristic Z-scan function for a thick sample is defined by the function,

$$F(\zeta, I) = \frac{1}{4} \ln \left(\frac{[(\zeta + l/2)^2 + 1] [(\zeta - l/2)^2 + 9]}{[(\zeta - l/2)^2 + 1] [(\zeta + l/2)^2 + 9]} \right), \quad (2)$$

where the parameters $\zeta = z/z_0$ and $l = L/z_0$ scale the scanning length and the cuvette length, respectively. The effective nonlinear index coefficient, n_2^{eff} , is then extracted from the fit of the Z-scan curves.

We use a refractive index of 1.43 [38], which agrees with our previous measurement of the dispersion and refractive index of 1-decanol [34]. Since n_2^{eff} is extracted only from the low-intensity Z-scan data, where the spectrum does not broaden significantly, Eq. (1) depends only on the refractive index at the pump wavelength. Given that the refractive index of 1-decanol varies by <1% throughout the measured range, treating it as wavelength-independent is justified for this analysis.

Figure 2(a) shows the low-intensity regime where we do not see SC, but the data follows a typical Z-scan shape. By fitting the data to Eq. (1), we determine the effective nonlinearity, $n_2^{\text{eff}}(I_0)$, at each intensity. The effective nonlinearity increases linearly as a function of peak intensity in the low-intensity regime. Therefore, Fig. 2(b) illustrates how in the low-intensity regime, we can

fit n_2^{eff} as a function of intensity to calculate $n_4 = 7.22 \times 10^{-35} \text{ m}^4/\text{W}^2$. Extrapolating the linear fit to the y-intercept yields the Kerr coefficient $n_2 = 1.87 \times 10^{-20} \text{ m}^2/\text{W}$.

Compared to some values of n_2 reported in the literature for CS_2 , a commonly used medium for supercontinuum generation, the n_2 of 1-decanol is lower by approximately an order of magnitude ($n_2(\text{CS}_2) \approx 2.8 \times 10^{-19} \text{ m}^2/\text{W}$) [39,40]. However, n_4 for CS_2 is reportedly $n_4 = -2 \times 10^{-35} \text{ m}^4/\text{W}^2$ at 800 nm [41], where the change of sign is related to the location of the third harmonic compared to the absorption bands of the medium [42]. Therefore, we have shown that 1-decanol exhibits a degree of optical nonlinearity on par with CS_2 without a high degree of toxicity. In fact, using the cited values, the effective nonlinear index of 1-decanol exceeds that of CS_2 above $2.8 \times 10^{15} \text{ W/m}^2$, which is the regime where we report SC generation.

Aside from our previous measurement [33], there are very few known measurements of the optical nonlinearity of 1-decanol. Ho et al. report $n_2 = 10.9 \times 10^{-20} \text{ m}^2/\text{W}$, measured using a picosecond (ps) laser centered at $1.06 \mu\text{m}$ [43], while Kuzyk and Dirk [36] obtain $n_2 \approx 622 \times 10^{-20} \text{ m}^2/\text{W}$ using a nanosecond (ns) laser centered at 1064 nm by converting the Kerr constant B_0 reported in Ref. [44]. Table 1 summarizes all known reports of n_2 for 1-decanol. It should be noted that the nonlinear phase imparted by the cuvette is small compared to that of the liquid, due to the thickness of the cuvette walls being much smaller than that of the liquid. Nevertheless, we do include the nonlinearity of the cuvette in our model [45].

Table 1. Reported Kerr coefficient n_2 values for 1-decanol.

Reference	Wavelength (nm)	Method	$n_2 (\text{m}^2/\text{W})$
This work	785	Closed-aperture Z-scan	1.87×10^{-20}
Our previous work [33]	785	Z-scan	6.8×10^{-20}
Ho et al. [43]	1064	ps OKE	10.9×10^{-20}
Kuzyk & Dirk [36]	1064	ns OKE [44] ^a	622×10^{-20}

^aOptically induced Kerr constant B_0 reported in Ref. [44] and converted to n_2 in Ref. [36].

Figure 3 shows the high-intensity, SC regime where we observe an intensity dependent thermal effect consistent with the generation of a spatial optical soliton [46]. In this regime, the data do not follow the Z-scan shape exhibited in Fig. 2(a). Because the Z-scan technique breaks down in this regime, we do not use it to calculate the nonlinear index of the medium. The breakdown of the fit begins at $>1 \times 10^{15} \text{ W/m}^2$ and becomes more apparent $>2 \times 10^{15} \text{ W/m}^2$. We argue that multi-photon absorption is not responsible for the disagreement with the Z-scan fit because we do not measure additional power loss through the cuvette at these high intensities.

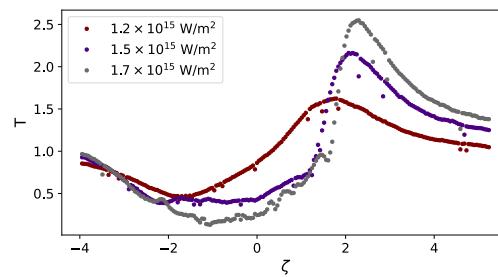


Fig. 3. High-intensity Z-scans. In this regime, the Z-scan fit (Eq. (1)) breaks down due intensity-dependent thermal effects. At these intensities, we observe supercontinuum generation near $\zeta = 0$.

3.2. Spectrum vs. axial position

Figure 4(a) illustrates the intensity of the spectrum as a function of wavelength and cuvette position. As the 1-decanol approaches the focus, the spectrum undergoes significant spectral broadening, creating a SC which is stable until the 1-decanol passes out of the focus and returns to its initial bandwidth. The WLC spans from approximately 450 nm to 950 nm, and the significant extension on the blue side of the spectrum at $\zeta = -2$ can likely be attributed to self-steepening [47], and we note that the upper limit of our spectrometer is approximately 1000 nm due to the silicon detector, thus limiting our measurements from extending further into the infrared. We also previously measured the absorption spectrum of 1-decanol and found that it is weakly absorbing beyond 900 nm [34].

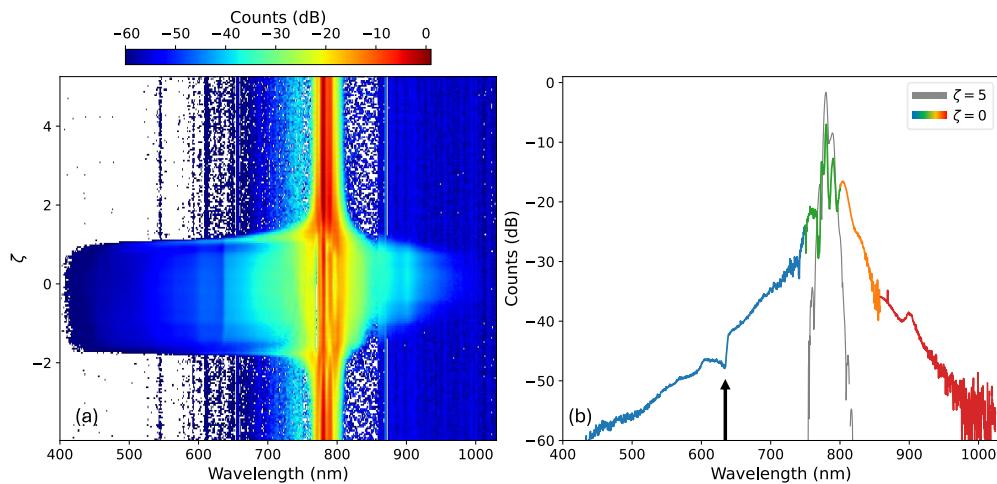


Fig. 4. (a) As the cuvette passes through the focus ($\zeta = 0$), the spectrum rapidly broadens into a white light continuum. (b) A lineout of the spectrum at the focus ($\zeta = 0$) and far from the focus ($\zeta = 5$). Colours indicate the bandwidth of each optical filter used in the measurement; arrow indicates position of the Raman loss.

Figure 4(b) shows lineouts contrasting the initial spectrum ($\zeta = 5$) to the SC ($\zeta = 0$). The peak intensity of the beam at the focus is estimated to be $3 \times 10^{15} \text{ W/m}^2$. The difference in energy between 785 nm and 636 nm corresponds to a Raman shift of -2984 cm^{-1} , which lies within the broad Raman response of 1-decanol centered at 2900 cm^{-1} [48], indicating Raman loss caused by the pump (black arrow) [49–52]. Apart from this localized feature, Raman effects do not dominate the supercontinuum generation process. Since we operate in the normal dispersion regime for 1-decanol, we do not generate the strong Raman frequency shift that is characteristic of anomalous-dispersion soliton dynamics. Furthermore, our femtosecond pump pulse is too short for efficient stimulated Raman cascade effects on the red side of the pump [53]. Therefore, we argue that the mechanism for supercontinuum generation is dominated by self-phase modulation and self-steepening. However at the high intensities shown in Fig. 3, additional nonlinear effects come into play and absorption, thermal lensing, self-focusing, and diffraction allow for extending the high-intensity interaction length, enabling more efficient SC generation via the generation of a spatial optical soliton [46,54].

3.3. Spectrum vs. radial angle

The radial distribution of the spectrum, as shown in Fig. 5, is such that all wavelengths in the supercontinuum are present near the center of the beam. However, while the visible wavelengths

remain present at greater radial distances, the infrared components do not. Furthermore, we do not observe significant spectral peak shifts off-axis.

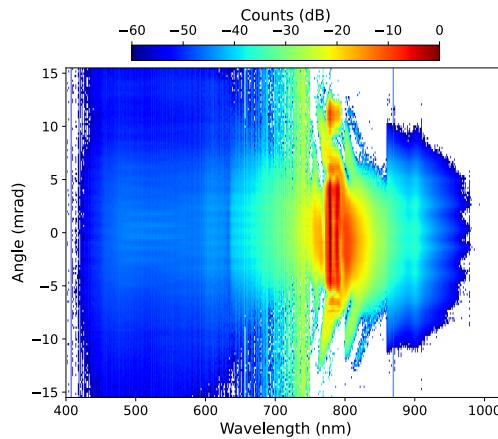


Fig. 5. The broad spectrum produced in 1-decanol as a function of radial angle from the center of the output beam. The visible portion exhibits a large divergence, indicative of the tight spatial confinement in the supercontinuum generation process.

The visible portion of the spectrum is relatively uniform and not significantly radially dependent over this divergence. This angle-wavelength distribution exhibits a different pattern than the X-wave amplification exhibited from four-wave mixing [55].

3.4. Temporal stability of the spectrum

Laser power fluctuations and material degradation would cause instability in the edges of the supercontinuum. As shown in Fig. 6, we measured the temporal stability of the SC over the main portion of the spectrum for 30 minutes. A benchmark of stability for SC spectra in liquids is found in [16] where the authors report a spectrum stable for over 70 hours using CS_2 in a liquid-core fiber. Because of the high boiling point of 1-decanol, we observe little evaporation of the sample after several weeks of repeated use and we expect similar longevity in our sample.

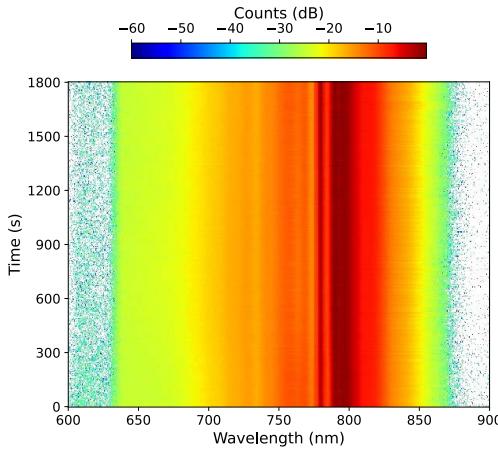


Fig. 6. The temporal stability of the most intense portion of the spectrum over 30 minutes, demonstrating little variation in the supercontinuum.

4. Conclusion

We show that it is possible to generate a broad white light continuum in 1-decanol that spans from 450 to 950 nm. Although the nonlinear Kerr coefficient is modest, we find that the higher-order nonlinearities significantly contribute to the measured phase shift. Not only is 1-decanol a much safer alternative to commonly used liquids such as chloroform (CHCl_3), carbon disulfide (CS_2), toluene (C_7H_8), and benzene (C_6H_6), but it demonstrates a nonlinear index on par with these liquids at high intensities. Additionally, we find that the supercontinuum is stable for extended periods of time, enabling prolonged study of strong-field physics. We expect that the comparable nonlinearity of 1-decanol to more toxic liquids makes it an attractive alternative for future nonlinear optics applications such as Kerr-instability amplification.

4.1. Data availability

- (1) TJ Hammond, "Data to replicate supercontinuum generation in 1-decanol," figshare (2014), <https://doi.org/10.5683/SP3/4HLC09>.

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Data availability. Data availability. Data underlying the results presented in this paper are available in Ref. [35].

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