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Systematic study of highly efficient white light generation in transparent materials using intense femtosecond laser pulses

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ABSTRACT We report the results of a systematic study of white light generation in different high band-gap optical media (BaF_2, acrylic, water and BK-7 glass) using ultrashort (45 fs) laser pulses. We have investigated the influence of different parameters, such as focal position of the incident laser light within the medium, the polarization state of the incident laser radiation and the pulse duration of the incident laser beam on the white light generation. Our results indicate that for intense, ultrashort pulses, the position of physical focus inside the media is crucial in the generation, with high efficiency, of white light spectra over the wavelength range 400–1100 nm. Linearly polarized incident laser light generates white light with higher intensity in the blue region than circularly polarized light. Ultrashort (45 fs) pulses generate a flatter spectrum with higher white light conversion efficiency than longer (300 fs) pulses of the same laser power. We believe that a flat response over a wide range of wavelengths in the continuum may be efficiently compressed for generation of sub-10 fs pulses.

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1 Introduction

The propagation of short pulses of intense laser light through an optical medium can lead to considerable temporal and spatial broadening that becomes mapped, in the frequency domain, into spectral broadening [1]. When the incident laser pulse is ultrashort, of femtosecond duration, the spectral broadening manifests itself in light that emerges out of the medium as a white disk surrounded by a distinct, concentric, rainbow-like pattern that is referred to as the conical emission; the central, low-divergence part of the output beam is referred to as the “white-light continuum” or “supercontinuum”; it can be readily separated either by placing an appropriate diameter aperture after the optical medium or making observations in the extreme far-field regime. Conversely, the conical emission can be selectively studied by masking the central white light portion. Supercontinuum generation has been observed in various media [1, 2]. Self-phase modulation (SPM) [3, 4], ionization-enhanced SPM [4, 5], four-wave mixing [3] and SPM enhanced by self-steepening of the incident pulse [6] are some of the mechanisms that were invoked in early studies to explain the generation of the white-light continuum, and the extent of its spectral width. But it is not unfair to state that the physics governing supercontinuum production is not properly understood despite its intrinsic interest and its utility in contemporary schemes for the generation of multigigawatt ultra-short, sub-5 fs pulses [7].

It is now generally accepted that supercontinuum generation is most likely a result of the interplay of a number of dynamical processes [2], such as self-focusing, group velocity dispersion, intensity clamping, self-steepening, anti-Stokes spectral broadening, filament fusion, filament breakup and competition between multiple filaments. Self-focusing clearly plays the role of the initiator of the sequence of processes that lead to white light generation [8–10]. When the laser power that is incident on a medium exceeds the critical power required for self-focusing, $P_{\text{cr}} = \lambda_o^2/2\pi n_o n_2$, a catastrophic collapse of laser energy occurs at a finite distance. n_o and n_2 denote, respectively, the linear and nonlinear refractive indices of the medium, and λ_o is the wavelength of laser radiation in vacuum. Experiments have shown that the power threshold for continuum generation coincides with the calculated critical power for self-focusing, in line with the proposal originally made by Bloembergen [11] to explain the white light continuum obtained using long-pulse (picosecond duration) laser light. In this model, self-focusing is stopped by avalanche ionization; the appearance of free electrons enhances SPM and gives rise to the continuum. In the short-pulse regime (femtosecond duration pulses), multiphoton excitation (MPE) has been shown to be an important mechanism of free-electron generation [12]. Furthermore, the band gap of the material exerts a strong influence on white light generation and self-focusing: there appears to exist a band gap threshold of 4.7 eV below which optical media do not generate a continuum, and above which the spectral width of the continuum increases with band gap [13]. It has been suggested that the enhancement of SPM by free electrons that are generated by MPE may be the primary mechanism of continuum generation [12, 13].

It is clear that self-focusing within a medium cannot proceed indefinitely. Hence, there is a need for a mechanism that arrests its collapse and, in condensed media; multiphoton ionization (MPI) may be such a process [13]. The ion-

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ization process places demands on the incident energy of the collapsing field; free-electron generation results in the production of plasma that, in turn, absorbs, defocuses, and spectrally blue-shifts the intense laser field. The combined effect of MPI and plasma defocusing limits further collapse of self-focusing and clamps the maximum intensity that can be accessed by the collapsing pulse. Thus, limiting of the maximum intensity is believed to be one of the dominant factors that determine the spectral extent of supercontinuum generation [14].

Recent experiments have offered indications that chromatic dispersion [12] might also contribute to limiting the spectral extent of supercontinuum generation. Normal group-velocity dispersion can give rise to a splitting of the incident laser pulse that may provide a balance to the self-focusing effect.

On the experimental front, there continues to be a paucity of information on self-guiding of femtosecond pulses and white light formation in bulk transparent media; there is a clear need for more systematic experimental studies in order that some insight begins to be developed into the complex physics that determines supercontinuum production. There are only a handful of reports on propagation, mostly in materials like fused silica [15–17]. Results of a study of supercontinuum formation in water and air has been recently published [2] that include experimental evidence for multiple filamentation obtained using a terawatt laser pulse, and concomitant interference effects in the spectral components of the white light that is generated as well as information on the dependence of the white light conversion efficiency on the length of the filament and the introduction of chirp in the incident laser beam. Direct visualization of multiple filaments has been achieved by means of a nonlinear two-photon fluorescence technique [18]. It is known that the white light spectrum is essentially highly asymmetric and may, thus, possess a disadvantage for pulse compression to generate sub-10 fs pulses. In this context there would, therefore, be a distinct advantage to develop schemes wherein one can generate white light spectra that has a flat response over a wide range of wavelengths so that subsequent compression can proceed efficiently.

We have recently initiated experiments on propagation studies using high intensity, ultrashort laser pulses through condensed media like barium fluoride and glass. A preliminary report has recently appeared on highly efficient white light generation in a 10 cm long barium fluoride crystal upon irradiation by 45 fs long pulses of 800 nm laser radiation with incident energy up to 1 mJ [19]. A nearly flat visible band along with the near-infrared region of the spectrum was generated in such experiments, spanning the wavelength region from 400 nm to 1100 nm. A minimum efficiency of ∼ 40%, an unexpectedly high value, was measured across the entire band for this process. Along with white light generation, multiphoton absorption induced fluorescence was also directly observed within the BaF_2 crystal. The highly coherent nature of the white light was demonstrated [19] by the formation of interference fringes that arise from multiple filaments that are created within 8 mm thick BK-7 glass upon its irradiation by a single, low-intensity laser beam employing line-focusing geometry.

We report here the results of a systematic study of white light generation from high band-gap materials, barium fluoride, acrylic, water and BK-7 glass upon their irradiation by focused, 800 nm, laser light. We probe the dependence of the intensity and spectral distribution of white light generation on physical focusing conditions, on the polarization properties of the incident light, and on laser pulse duration. The results that are presented may be of use in optimizing supercontinuum production using various high band gap materials and, it is hoped, will stimulate much-needed theoretical work.

2 Experimental setup

Ultrashort laser pulses used in the present series of experiments were generated using a conventional chirped pulse amplification (CPA) system. Briefly, the system comprised an oscillator that delivered an 88 MHz pulse train with an average mode-lock output power of 500 mW. The full width half maximum bandwidth of the oscillator pulses, as measured with a spectrometer, was typically ∼ 55 nm. The pulse train from the oscillator was directed into an amplification system consisting of a pulse stretcher, a pulse picker to slice the input pulse train to 1 kHz, an amplifier comprising a Ti:sapphire crystal that was pumped by a 1 kHz Q-switched Nd:YLF laser, followed by a pulse compressor. The pulse made nine passes in the multipass amplifier in a ring configuration. After compression we obtained pulses of 45 fs duration with output energy of up to 1 mJ, at 1 kHz repetition rate; the corresponding bandwidth was measured to be typically ∼ 28 nm. The beam diameter was 3 mm. The dependence of white generation on laser pulse duration is also reported using a second Ti:sapphire, CPA-based laser with 300 fs pulse duration and 10 mJ energy at a repetition rate of 10 Hz. The output beam diameter in this case was 10 mm prior to focusing. Besides, the full width half maximum bandwidth of the oscillator pulses for this laser system was typically ∼ 28 nm; after the amplifier, the value of the bandwidth was measured to be ∼ 15 nm.

In a typical experiment, the beam from either the 45 fs laser or the 300 fs laser was focused on the samples (7.5 cm long barium fluoride crystal, 5 cm long acrylic block, 1 cm width BK-7 glass plate, and 5 cm cell containing deionized water) and the white light that was produced was detected so as to deduce values of conversion efficiency. We measured the laser energy that was incident on the high band gap material under study, using an energy meter with a nearly flat and wide spectral response that covered the range 400–1100 nm. The same energy meter was also used to measure the energy of the entire white light continuum. In the present series of experiments, the ratio of the latter to the former was taken to be the white light conversion efficiency. By changing the parameters of incident light, such as using lenses of a variety of focal lengths (5–30 cm) and optimizing the position of focus inside the media, we also measured white light spectra using a fibre-optic-coupled spectrometer. The efficiency of the grating used in our spectrometer was in excess of 30% over the entire spectral range, with efficiency in excess of 50% over the range 580–1050 nm. The spectrometer was also used to measure the spatial distribution of the spectral properties of the white light beam. By scanning a 500 μm wide slit along the x and y directions, we confirmed that there was no spatial chirp. This is important as, otherwise, the longer wavelength components of the white light might be underestimated vis-à-vis the shorter wavelength components by virtue of the former undergoing somewhat larger diffraction and, hence, larger divergence. The silicon charge coupled device that was used as the detector in the spectrometer had maximum spectral sensitivity at 550 nm; its sensitivity was above 50% of the maximum value over the range 400–750 nm, with a monotonic decrease as the wavelength increased beyond 750 nm.

Polarization dependent measurements were also conducted in our experiments. Circularly polarized light was obtained by passing the linearly polarized laser output through a quarter-wave plate. The extent of elliptical polarization is defined by the ellipticity parameter, $\varepsilon = (E_x/E_y)$, and in our experiments circular polarization implies a value of ε that lies between 0.9 and 1.0.

3 Results and discussion

We discuss here the results of white light generation in BaF_2, BK-7 glass, acrylic and water. We first focus attention on our experiments using laser pulses of 45 fs duration. Such pulses undergo temporal broadening when propagating through long length medium, reducing the light intensity. In our measurements, we have optimised, for a lens of given focal length, the location of focus inside the sample so as to obtain the flattest possible spectrum of white light with high conversion efficiency. Figure 1 shows some typical white light spectra obtained at different laser focus positions within the BaF_2 crystal. The incident laser power employed in this measurement was ~ 3000 times that of P_{cr} in BaF_2 ($P_{cr} = 3.9$ MW). Most previously reported work has been conducted using power levels that are close to P_{cr} where rings and conical emission constitute a significant portion of the total output signal. At the very high power levels used in our measurements, it is the central white light portion that overwhelmingly dominates the total signal output [19]. A lens with 30 cm focal length was used to acquire data shown in Fig. 1. When the laser beam was focused at a position that lies 1 cm from the front face of the crystal we obtained a nearly flat spectra, and high efficiency, with no damage to the medium. When the focus position was altered such that it was located near the centre of the 7.5 cm long crystal, the intensity of the spectra was found to lessen in the region 400–500 nm. When the laser beam was focussed well inside the medium (at 5.5 and 6.5 cm from the front face of the crystal) the intensity in the spectral region from 400–600 nm was significantly reduced. These observations can be rationalized by considering the fact that dispersion broadens the pulse duration of the laser, thereby reducing its intensity. To confirm this, we focussed the laser beam very close to the front face (locating the focal point only 0.3 cm inside the crystal) and confirmed that the broadest white light spectrum was, indeed, obtained under these conditions. However, we observed distinct damage to the crystal face under these very high intensity conditions.

In Fig. 2 we show white light spectra from BaF_2 that were obtained using different focal length lenses while keeping the laser energy constant. In the case of the shortest focal length (5 cm) lens we obtained the highest conversion efficiency, 80%, but this was accompanied by damage to the medium. Longer focal length lenses yielded somewhat lower conversion efficiencies, typically 66% and 68% for 10 cm and 30 cm lens, respectively. These measurements were conducted by collecting the entire white light generated from the medium by placing the energy meter in close proximity to the material under study. There was no damage to the crystal under these circumstances. We also investigated white light generation in other media, such as water, acrylic and glass and some typical spectra that we measured are shown in Fig. 3. In the case of acrylic, we observed nearly flat white light spectra, very similar to that observed in BaF_2. Our measurements showed that for a given input laser intensity, the white light intensity that we routinely obtained in water was higher than that in glass. This difference might be due to the different sample lengths

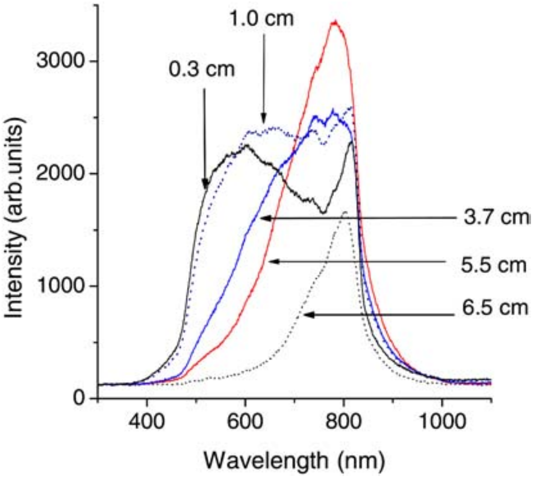

FIGURE 1 White light spectra obtained upon irradiation of 7.5 cm long BaF_2 crystal with the physical focus located at different distances within the crystal. A 30 cm lens was used for all measurements and the indicated distances are from the front face of the crystal. The incident laser energy was 600 μJ in each case. The vertical scales of the different spectra shown are not relative (see text for discussion of white light generation efficiencies under different conditions)

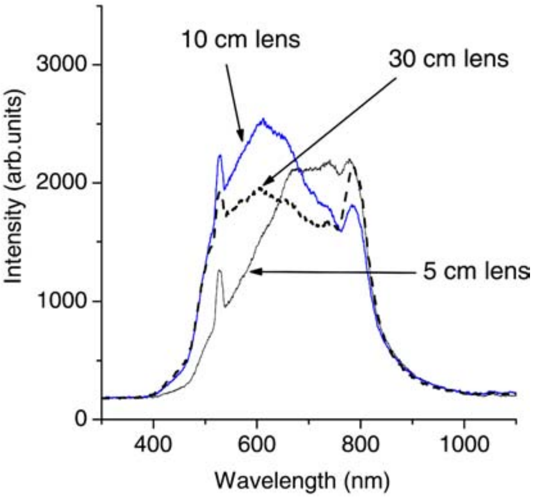

FIGURE 2 White light spectra obtained from BaF_2 using different focal length lenses, with the incident laser energy kept constant at 600 μJ

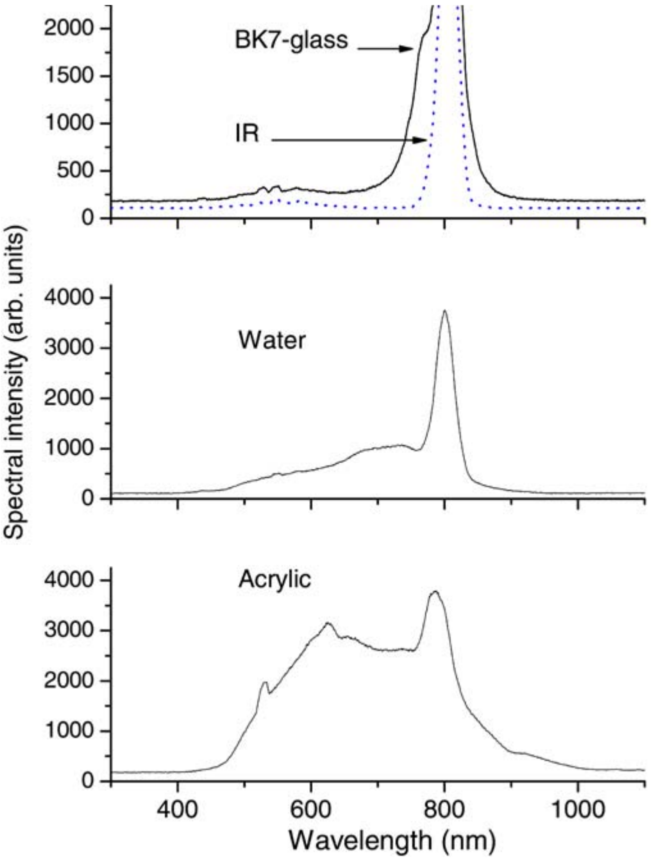

FIGURE 3 White light spectra obtained from water, acrylic and BK-7 glass with a 30 cm lens and incident laser energy of 600 μJ. The curve denoted IR depicts the spectrum of the incident laser light

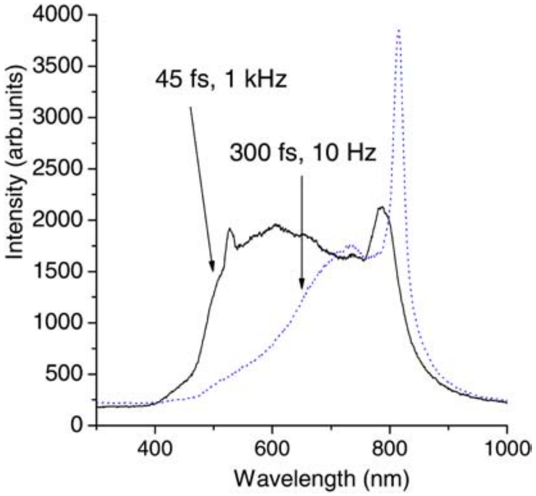

FIGURE 4 Comparison of white light spectra obtained from BaF_2 using the same incident laser power, with 45 fs pulse duration at a repetition rate of 1 kHz as well as 300 fs pulse duration at 10 Hz repetition rate

employed in our experiments, even though both these media satisfied the band gap criteria mentioned above. As noted earlier, measurement of the spatial distribution of the white light spectra obtained in our experiments offered no indications of spatial chirp.

We have also carried out experiments to investigate the dependence of white light generation on different pulse durations and on the polarization of the incident laser beam. The pulse duration dependence was carried out using two different lasers, with durations of 45 fs and 300 fs, using the same incident laser power. Figure 4 shows the white light spectra obtained using BaF_2 at different pulse durations with nearly the same incident laser power. Clearly, there is a marked difference in the spectra of white light that is generated using the two different pulses, particularly in the blue region of the spectrum. In the case of the 45 fs pulse, the white light spectrum is broad, and conversion appears to be better than in the case of 300 fs pulses. The above difference may be rationalized using the fact that SPM (Kerr nonlinearity) is known to be inversely proportional to the pulse duration [1]. Furthermore, free electron generation is expected to contribute to SPM, particularly enhancing the blue spectral region [12]. Thus, our measurements using pulses of two different durations seem to indicate that the free electron contribution to SPM is larger in the case of 45 fs pulses than for longer (300 fs) pulses. If we increase the laser energy in the case of 300 fs pulses, we observed severe damage to the medium under study whereas no damage was found with 45 fs pulses at incident energies as large as 1 mJ.

We now discuss our results using linearly and circularly polarized laser beams of 45 fs duration. Figure 5 depicts some typical spectra. Under conditions where the same energy (450 μJ) is employed, the signal intensity of white light was found to be higher for linearly polarized light compared to that obtained when circular polarization was used. This observation is rationalized on the basis of both MPE-enhanced SPM and SPM processes. In the case of femtosecond pulses, free electrons produced due to MPE enhance SPM, and contribute towards white light generation. Moreover, multiphoton effects are known to be dependent on the state of polarization of the incident light. Lambropoulos [20] pointed out that the effect of light polarization on the multiphoton ionization of atoms is related, in a general sense, to the effect of field correlations of multiphoton processes. Both effects arise from the fact that the vectors of the radiation field affect, in nonlinear fashion, the transition amplitudes for multiphoton processes. The nonlinearity in the amplitude of the radiation field leads to ionization rates that depend on the correlation functions of the field, and not just on the absolute value of the field amplitude. When the circular polarization vector $E_x + \mathrm{i}E_y$ is inserted in the expression for the transition amplitude, cross products of matrix elements involving the orthogonal components E_x and E_y occur, and these lead to the dependence of the ionization rate on the polarization state of the incident light field. Sandhu et al. [21] have shown that supercontinuum generation in water, using 100 fs pulses, is reduced for circularly polarized light. Petit et al. [22] have also shown that multiphoton ionization is less efficient when a circularly polarized laser beam is used. Other reports have theoretically shown that the effective ionization energy for circularly polarized beams is higher than for linearly polarized beam [23]. Recently, Fibich et al. have theoretically shown that multiple filamentation caused by self-focusing is suppressed for circularly polarized laser beam in a Kerr medium [24].

In gas-phase studies on strong field molecular dynamics conducted in our laboratory, considerable evidence has been

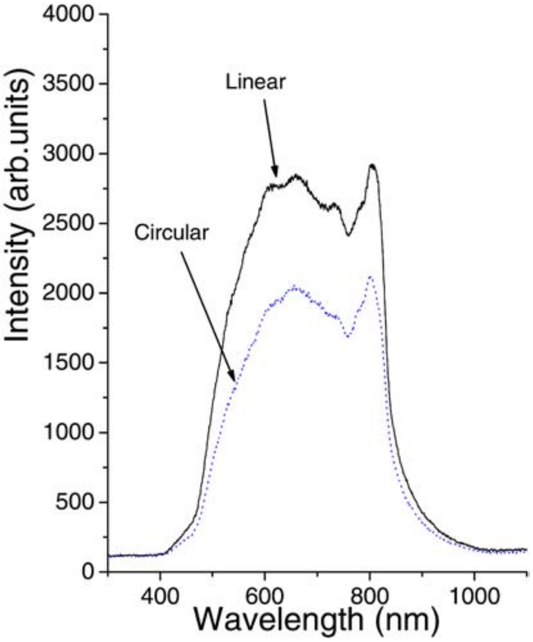

FIGURE 5 White light spectra from BaF_2 using linearly and circularly polarized light, keeping the incident energy constant at 450 μJ

accumulated in recent years on how the efficacy of strong-field multiphoton and field ionization is significantly reduced when linearly polarized light is replaced by circularly polarized light [25, 26].

In the case of cubic materials it is known that the third order susceptibility tensor contains off-diagonal elements, as a result of which the two-photon absorption (TPA) exhibits dependence on the polarization state of the light. In a single beam experiment it has been shown that the TPA coefficient is not the same for linear and circularly polarized light [27]. The polarization dependence of the multiphoton absorption coefficient can be obtained from corresponding higher-order nonlinearities. We now discuss how SPM processes contribute towards the polarization dependence of white light generation. BaF_2 is a cubic crystal with inverse symmetry; it is optically isotropic and does not show linear birefringence with optical field. However, when the crystal is subjected to a sufficiently intense optical field, SPM and cross phase modulation (XPM) [28, 29] may give rise to the field-induced change in the nonlinear refractive index, whose magnitude is intensity dependent. The presence of XPM gives rise to a nonlinear coupling between orthogonal field components. The nonlinear refractive index change, in general, is unequal for two orthogonal crystal axes since it depends both on the laser intensity and its polarization.

Spectra in Fig. 5 show that the blue side is broad and intense for the linearly polarized beam compared to that obtained with a circularly polarized beam. This might be rationalized by considering the difference of interaction intensities at which those spectral components were generated. Generation of the blue spectrum requires higher intensity than that of light in the central part of the overall output spectrum. Therefore, the variation of the white light intensity with laser polarization becomes most significant in the blue region of the spectrum.

In the context of data that we have presented, it is of interest to note the results of numerical simulations that have recently been carried out to study the polarization dynamics of femtosecond laser pulses propagating in air [30]. In these simulations, significantly more supercontinuum production was obtained when linearly polarized light was used than with equally intense, circularly polarized light. It was noted that supercontinuum generation depends strongly on the incident laser power and, as the critical power for circularly polarized light is 1.5 times that for linear polarization, the rationalization offered for the results of the simulations was in terms of circularly polarized pulses being 'weaker' [30]. Making a direct comparison between our experimental results and those of the numerical simulations is not straightforward as the computations are based on solutions of the nonlinear Schrödinger equation (NLSE). Recent work has shown the serious limitations of the ordinary NLSE approach in situations involving propagation of femtosecond-duration, intense laser pulses through media such as sapphire [31] and air [32]. As noted above, our experiments were conducted in a regime that ensured that incident laser powers, for both types of polarization, were two orders of magnitude in excess of the critical power for self-focusing. This is a regime where there remains an acute paucity of theoretical work.

4 Summary

We have conducted a series of systematic measurements on white light generation in different high band-gap media (BaF_2, acrylic, water and BK-7 glass) with a view to gaining information on how conversion efficiencies and spectral features are determined by parameters that can be easily controlled by experimentalists. The parameters that we have explored in this context are the focal position of the incident laser light within the medium, the polarization state of the incident laser radiation and the pulse duration of the incident laser beam. Our results indicate that (i) for short femtosecond pulses (45 fs), position of focus inside the media is crucial in the generation of flattest white light spectra with high efficiency, (ii) linearly polarized light generates white light with higher intensity in the blue region than circularly polarized light and (iii) short (45 fs) pulse generates flat spectra and with higher white light conversion efficiency than long (300 fs) pulse. It is hoped that these systematic studies will stimulate much needed theoretical work that will facilitate insights to be developed into the complex dynamics that govern white light generation in condensed media.

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