Toward an understanding of white-light generation in cubic media—polarization properties across the entire spectral range

Ivan Buchvarov,[1,2] Anton Trifonov,[1] and Torsten Fiebig[1,*]

[1]*Eugene F. Merkert Chemistry Center, Boston College, 2609 Beacon Street, Chestnut Hill, Massachusetts 02467, USA*
[2]*Permanent address: Faculty of Physics, University of Sofia, Bulgaria*
[*]*Corresponding author: fiebig@bc.edu*

Received October 20, 2006; revised February 28, 2007; accepted April 2, 2007;
posted April 6, 2007 (Doc. ID 76282); published May 15, 2007

We present an extensive investigation of the polarization properties of a femtosecond-laser-induced white-light (WL) continuum generated in a cubic crystal (CaF_2). The WL spectrum and threshold energies have been examined for input polarizations with various degrees of ellipticity. For linear input polarization, the WL spectrum shows strong depolarization around the input wavelength, while the preservation of the input polarization is pronounced toward the blue spectral region. The observed depolarization effect has been elucidated in the framework of current models for WL generation. © 2007 Optical Society of America

OCIS codes: 320.7110, 320.2250, 190.5940, 260.5430, 300.6530.

The interaction of intense laser pulses with transparent media can result in vast spectral broadening, ranging from the infrared to the ultraviolet spectral region. This continuum or white-light generation (WLG) is a well-established phenomenon [1,2]. Femtosecond-laser-induced white light (WL) has been the source of ultrashort coherent radiation for numerous applications: time-resolved broadband pump–probe spectroscopy [3], optical pulse compression [4], and optical parametric amplification [5].

In spite of its importance for pure and applied sciences, the physical mechanism of WLG in solids is not completely understood. It is widely assumed that several nonlinear processes contribute to WLG: (i) Self-phase modulation (SPM) [1], (ii) optical "shock-wave" formation [6–8] due to self-steepening, (iii) space–time focusing [9], and (iv) plasma generation by multiphoton ionization [10]. In addition to these processes, it was pointed out that chromatic dispersion plays a role as well [11]. The WLG is triggered by a "catastrophic beam collapse" when the input pulse intensity is above the threshold for self-focusing (SF) in certain materials [2]. This extreme SF and the concomitant increase of the light intensity is suppressed by multiphoton absorption and a negative lens effect originating from a low-density plasma. The dynamic interplay between the various contributing processes leads to the formation of optical filaments in the medium [12].

A subject of frequent debates is not only the broad blueshifted wing of the WL spectrum but also its polarization properties [1,13–16]. Since the discovery of WLG, it has been commonly assumed that the polarization of the generated WL in isotropic condensed media inherits the polarization of the input laser pulse [1,13]. Until now, investigations of the WL polarization were either carried out in an integrated fashion, i.e., over the entire spectrum [1,13] or in a very narrow spectral range close to the wavelength of the input pulse [14,15]. The knowledge of the polarization properties of all spectral components of the WL will have a profound impact on both applications and theoretical modeling [15] of the WLG.

In this Letter, we report our extensive investigations of the polarization properties of a femtosecond-laser-induced WL continuum in a cubic crystal (CaF_2). In the case of linear input polarization, we observed a strong spectral dependence of the WL polarization state. In the spectral vicinity of the input pulse, depolarization is dominant whereas the blue-wing spectral region is characterized by a large degree of polarization preservation. We also demonstrate that the WL cutoff frequency of the blue wing, the threshold energy for WLG, and the WL spectral intensity distribution, depend strongly upon the ellipticity of the input pulse polarization.

The experiments were performed with a Ti:sapphire system (CPA 2010 Clark-MXR) generating 180 fs pulses at 775 nm with a 1 kHz repetition rate. A small fraction of the laser output was focused into a 3 mm thick CaF_2 plate by a lens ($f=100$ mm, producing a spot with a diameter of 32 μm). To avoid optical damage, the plate was rotated. The beam path was parallel to one of the fourfold crystal axes [001]. The initiation of the WLG was achieved by adjusting the pulse energy using a variable neutral density filter. The beam diameter on the focusing lens was 3.5 mm at $1/e^2$. The performance of the designed WL generator was described previously [3,17]. A single filament WL was obtained by increasing the pulse energy slightly above the SF threshold. The WL signal was collimated using a high-quality parabolic mirror. The polarization of both the input beam and the WL was determined and analyzed by two Glan polarizers (extinction ratio $<10^{-4}$). Monitoring of the WL output signal was done using a commercial spectrograph. The error caused by the polarization-dependent response of the spectrograph was in the range of 10%.

In the case of linear input polarization, the outgoing beam from the first polarizer was focused into the CaF_2 plate. The polarization of the generated WL continuum was examined by measuring the trans-

0146-9592/07/111539-3/$15.00

mitted light through the analyzer at perpendicular $I_{\perp}(\lambda)$ and at parallel $I_{\parallel}(\lambda)$ orientations with respect to the input polarizer as a function of the wavelength λ. Therefore the input polarization retained in the process of continuum generation can be presented by the polarization ratio $\rho(\lambda)=I_{\parallel}/I_{\perp}$. Hence, the larger the observed changes of the WL polarization (here called "depolarization") the smaller the resulting values of ρ, e.g., for the input pulse $\rho>10^4$. The WL spectrum and its polarization ratio $\rho(\lambda)$ are shown in Fig. 1. The WL spectrum is characterized by symmetric broadening ($\sim$2000 cm^{-1}) around the input wavelength followed by a broad blueshifted pedestal with an intensity ratio of 10^{-3} to 10^{-4} (with respect to the input intensity at 775 nm). As exhibited in the figure, the spectral broadening extends all the way into the ultraviolet (≤300 nm). The $\rho(\lambda)$ curves in the figure illustrate the substantial wavelength dependence of ρ. The sharp peak ($\rho>100$) at 775 nm is surrounded by a pronounced WL depolarization ($\rho<3$). In contrast, the blue region of the WL (300–650 nm) shows only minor depolarization ($\rho>10$), and ρ rises monotonically toward the blue edge of the spectrum. Upon increasing the input pulse energy, ρ decreases throughout the far blue part of the spectrum (500–300 nm), as shown in Fig. 1. In principle, the observed depolarization may be attributed either to interactions of the propagating pulse with the plasma [16] or to the anisotropy of the nonlinear refraction in cubic crystal (described by the $\chi^{(3)}$ [18,19]). The $\chi^{(3)}$ tensor of a cubic material is intrinsically anisotropic, i.e., it contains some nonzero off-diagonal elements. The $\chi^{(3)}$ anisotropy leads to (i) the generation of an orthogonal polarized wave and thus to changes of the polarization of a monochromatic input wave (referred to as the self-action polarization effect [18–20]) and (ii) the alteration of the polarization of a monochromic wave by interaction with a stronger wave with different frequency, when both waves are polarized in the same direction. The latter effect is known as the "pump–probe nonlinear anisotropic polarization effect" [20]. For example, the strong wave at 775 nm will affect the polarization of the weaker waves in the blue spectral region. In terms of nonlinear refraction, both effects (i) and (ii) can be considered as nonlinear anisotropic birefringence (NAB), and its magnitude depends on the crystal orientation (with respect to the input polarization). For cubic crystals, the NAB vanishes at eight distinct angles ($\alpha=m\pi/4$, $m=0,1,\ldots$, where α is the angle between the crystal orientation [100] and the input polarization) [18–20]. Hence, the relative contribution of the polarization change caused by the anisotropy of $\chi^{(3)}$ can be evaluated by measuring ρ as a function of the angle α. Indeed, we have found eight angles ($\alpha=m\pi/4$, $m=0,1,\ldots$) for which the input polarization is maintained, i.e., $\rho>10^3$ while at the other orientation angle α, the observed polarization change is substantial with maxima at $\alpha\approx\pm13°+m\pi/2$, $m=0,1,\ldots$. The lack of depolarization at $\alpha=0°$ (Fig. 1 inset) clearly shows that under our experimental conditions, the influence of the plasma on the WL depolarization can be neglected.

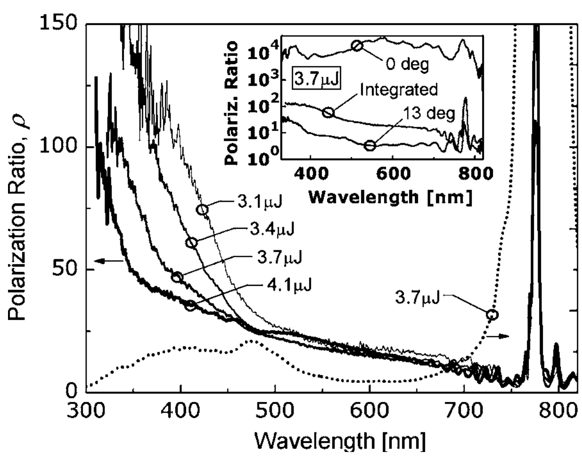

Fig. 1. Polarization ratios (solid curves) and WL spectrum (dotted curve) generated with linearly polarized input pulses at various input energies. The WL threshold energy was 2.2 μJ. The inset shows the polarization ratios at $\alpha=0°$, 13° and integrated over a full rotation period, respectively. The rotation of the CaF_2 plate was replaced by a translational motion only for the measurements at $\alpha=0°$ and 13°.

Most striking is that $\rho(\lambda)$ increases toward the blue edge of the WL spectrum. Obviously this result cannot be explained by a simple intensity-dependent birefringence model [14], which predicts that ρ is proportional to λ^2 [21]. We also found that incorporating the group-velocity mismatch into this model does not reproduce the observed wavelength dependence of ρ. However, spectral dependence of the WL polarization can be elucidated in the framework of femtosecond-pulse shape propagation dynamics in condensed media.

Guided by the experimental results and the recent theoretical description of WLG [7,11,22], we propose the following mechanism. We assume that different spectral components of the WL are generated at different positions along the propagation pathway of the pulse through the medium. The fate of the polarization of each component is determined by its corresponding NAB and its interaction length in the WL filament. The multiphoton absorption and plasma defocusing weaken the trailing edge of the pulse. At this stage, the beam diameter shrinks and approaches its minimum. A sharp peak appears at the front edge that results in spectral broadening around the input laser line [7]. Subsequently, space–time focusing and self-steepening create a steep back edge, which results in a broad blue wing of the WL spectrum [6,7]. The shock-wave profile is formed at a certain distance from the filament starting point. Therefore the effective length of interaction of the WL spectrum components with the most intense part of the pulse at 775 nm decreases toward the blue part of the spectrum, i.e., more pronounced depolarization (through the NAB) will be observed for the WL spectrum around the input wavelength, while minor depolarization occurs in the blue wing. Furthermore, it is important to emphasize that the sharp peak of ρ at the input wavelength is due to the residual input pulse energy surrounding the filament.

When a quarter-wave plate (at 775 nm) was placed between the input polarizer and the CaF_2 plate, the ellipticity of the input polarization was varied by rotating the plate with respect to the polarizer's axis. The WL intensity was measured as a function of the input degree of ellipticity. The obtained WL spectra (Fig. 2) can be grouped into two types of spectral distributions, representing both linear and circular input polarizations, respectively. The Fig. 2 (inset) shows the WL intensity at 500 nm as a function of the input pulse energy for linear and circular input polarizations. Clearly, the WLG threshold depends strongly on the polarization of the input pulse [23].

In conclusion, we have presented, to the best of our knowledge, the first systematic investigation of the white-light polarization properties in an isotropic medium with cubic crystal structure across the entire anti-Stokes spectral range. The ellipticity of the input polarization determines the threshold for WLG as well as the cutoff wavelength and the intensity of the anti-Stokes pedestal. The observed WL depolarization in the case of linear-polarized input exhibits a strong spectral dependence, and its value decreases toward the blue wing of the spectrum. This experimental observation was interpreted as an optical shock-wave formation in the presence of nonlinear anisotropic birefringence. The results presented here provide an important aid to experimentalists who seek to employ the white light as a coherent light source with well-defined polarization properties. In addition, the polarization properties present a unique experimental tool for verifying theoretical hypotheses about the WLG process.

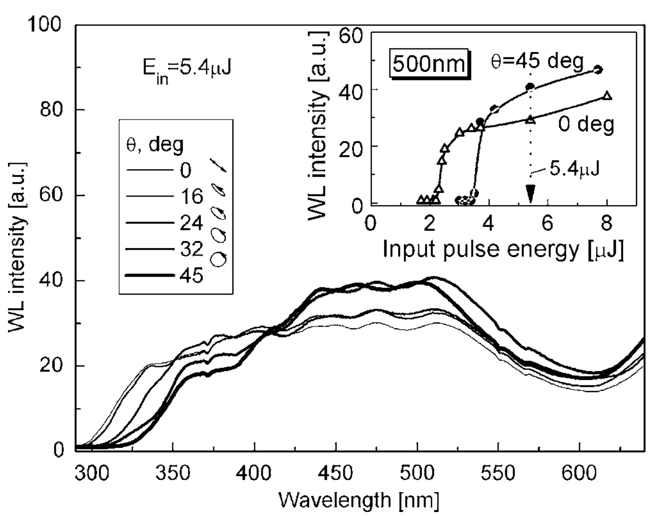

Fig. 2. WL spectra at various elliptical input polarizations, corresponding to different wave-plate rotation angles θ. The inset shows the WL intensity at 500 nm versus the input pulse energy for linear (triangles) and circular (circles) input polarizations.

We gratefully acknowledge useful discussions with A. Couairon (École Polytechnique, France) and V. Zanza (ENEA Centro Ricerche Frascati, Italy). This work was supported by the National Science Foundation (grant CHE-0521503).

References

1. R. R. Alfano, ed., *The Supercontinuum Laser Source* (Springer, 1989).
2. A. Brodeur and S. L. Chin, Phys. Rev. Lett. **80**, 4406 (1998).
3. M. Raytchev, E. Pandurski, I. Buchvarov, C. Modrakowski, and T. Fiebig, J. Phys. Chem. A **107**, 4592 (2003).
4. A. Baltuska, T. Fuji, and T. Kobayashi, Opt. Lett. **27**, 306 (2002).
5. P. Tzankov, T. Fiebig, and I. Buchvarov, Appl. Phys. Lett. **82**, 517 (2003).
6. A. L. Gaeta, Phys. Rev. Lett. **84**, 3582 (2000).
7. H. Ward and L. Berge, Phys. Rev. Lett. **90**, 053901 (2003).
8. V. P. Kandidov, O. G. Kosareva, I. S. Golubtsov, W. Liu, A. Becker, N. Akozbek, C. M. Bowden, and S. L. Chin, Appl. Phys. B **77**, 149 (2003).
9. J. E. Rothenberg, Opt. Lett. **17**, 1340 (1992).
10. N. Bloembergen, Opt. Commun. **8**, 285 (1973).
11. M. Kolesik, G. Katona, J. V. Moloney, and E. M. Wright, Appl. Phys. B **77**, 185 (2003).
12. A. Couairon, L. Sudrie, M. Franco, B. Prade, and A. Mysyrowicz, Phys. Rev. B **71**, 125435 (2005).
13. I. Golub, Opt. Lett. **15**, 305 (1990).
14. K. Midorikawa, H. Kawano, A. Suda, C. Nagura, and M. Obara, Appl. Phys. Lett. **80**, 923 (2002).
15. M. Kolesik, J. V. Moloney, and E. M. Wright, Phys. Rev. E **64**, 046607 (2001).
16. A. K. Dharmadhikari, E. A. Rajgara, and D. Mathur, Opt. Lett. **31**, 2184 (2006).
17. P. Tzankov, I. Buchvarov, and T. Fiebig, Opt. Commun. **203**, 107 (2002).
18. D. C. Hutchings, Opt. Lett. **20**, 1607 (1995).
19. D. C. Hutchings and B. S. Wherrett, Phys. Rev. B **52**, 8150 (1995).
20. Y. P. Svirko and N. I. Zheludev, *Polarization of Light in Nonlinear Optics* (Wiley, 1998).
21. $\rho(\lambda) \sim [\sin^2(\Delta\phi_{\mathrm{NL}}(\lambda)/2)\sin^2(2\alpha)]^{-1} \sim (\Delta\phi_{\mathrm{NL}}(\lambda)/2)^{-2} \sim [(2\pi/\lambda)L\Delta n_{\mathrm{brn}}(\alpha)]^{-2} \sim \lambda^2$, where $\Delta\phi_{\mathrm{NL}}(\lambda)$ is the nonlinear phase shift, λ is the wavelength, L is the interaction length, and Δn_{brn} is the effective nonlinear refraction index of the induced birefringence.
22. A. Couairon, E. Gaizauskas, D. Faccio, A. Dubietis, and P. Di Trapani, Phys. Rev. E **73**, 016608 (2006).
23. The obtained ratio between the WL energy thresholds for circular and linear polarization is $\sim 3/2$, which matches the ratio between the corresponding nonlinear indices of refraction (n_2). It is relevant for our experimental observations and also that for other groups that WLG threshold power is slightly above the SF threshold, i.e., it is also proportional to $\sim 1/n_2$.