RAPID COMMUNICATION

Low temperature enhancement of alignment-induced spectral broadening of femtosecond laser pulses*

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The propagation of femtosecond laser pulses in N_2-filled hollow fibers is studied both theoretically and experimentally. The laser pulse aligns the N_2 molecules and changes the refractive index , which meanwhile modulates the spectrum of the pulse in turn. The dependence of the spectral modulation on the gas temperature is investigated. We find that both spectral broadening and frequency red-shift are enhanced at low temperature. The degree of enhancement is found to be dependent on the pulse duration. Based on our findings, we propose a method for femtosecond pulse spectral broadening and few-cycle pulse generation via the molecular alignment.

Keywords: field-free molecular alignment, femtosecond phenomenon, ultrafast nonlinear optics

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1. Introduction

Since the 1970s, the molecular response of rotational coherence to pulsed excitations and its contribution to the nonlinear refractive index have been studied.[1,2] In 1995, Friedrich *et al.*[3] and Seideman[4] used time-dependent quantum mechanics to analyze this molecular response theoretically. The effect of molecular alignment was first proposed then to denote molecules becoming spatially aligned along the polarization direction when they are rotationally excited by a laser of moderate intensity. The alignment is divided into two types according to different relationships between the aligning pulse duration and the rotational period of molecules. The adiabatic alignment dominates when the pulse duration is longer than the rotational period[5] and the field-free alignment occurs when the pulse duration is much shorter and the alignment continues even after the effect of the laser has ceased.[6,7] The contribution of molecular alignment, especially the field-free alignment, to the nonlinear refractive index has also been studied. By introducing a delayed Raman response function $R(t)$ and analyzing the spectral modulation, Nibbering *et al.*[8] investigated how the refractive index is influenced by the rotational excitation. Their results demonstrated the time-delayed characteristics of the index modulation by molecular alignment. In 2002, Bartels *et al.*[9] provided a clear picture describing how the molecular index is modulated after the molecules are aligned by a linearly polarized laser pulse.

Subsequently, there have been many reports focused on this alignment-induced modulation and its applications. Generally, there are two types of applications. The first mainly focuses on the influence of the aligned molecules, which have already been pre-aligned by an aligning pulse, on a second delayed pulse. Based on this model, our group developed a method to generate widely tunable, few-cycle laser pulses.[10,11] Some additional studies have also been carried out with the combined effect of the filamentation process and this pre-induced index modulation.[12–16] The second type of application concerns how the molecular alignment influences the aligning pulse itself. Nisoli *et al.*[17] once observed that by using an input pulse with a duration of 20 fs, the spectral broadening through N_2 was mainly induced by the third-order nonlinear polarization. However, when the pulse duration was lengthened to 140 fs, there was a considerable spectral modulation and a clear red-shift was observed. They attributed this pulse-duration-associated difference to the time-delayed characteristics of the alignment-induced modulation. The propagating distance was only several centimeters in the work of Nisoli *et al.*[17] While in this paper, with the help of a hollow fiber and a low temperature environment, we study how to optimize this real-time spectral modulation of the aligning laser pulse and make it applicable for the few-cycle femtosecond pulse generation.

The molecular alignment is enhanced at low temperature. This is why molecular beams at low temperature have been used in orientation-dependent ionization[18,19] and high harmonic generation.[20–22] The optimal pulse duration and molecular temperature for index modulation (Δn),[23,24] spectral red-shift ($\mathrm{d}n/\mathrm{d}t$)[23,24] and spectral broadening ($\mathrm{d}n^2/\mathrm{d}t^2$)[23] have already been investigated theoretically. However, in all those reports, the effect of a long distance propagation has not been considered. It is also unclear how broadly the spectrum can be expanded and to what extent the center wavelength can be tuned. Herein, the spectral mod-

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ulation of the aligning femtosecond laser pulses themselves, when they propagate through an N_2-filled hollow fiber at different temperatures, is studied both theoretically and experimentally. We find that the spectrum of an initial chirped 100-fs pulse can be broadened considerably by the N_2 molecular modulation at low temperature. The output spectrum is wide enough to support a few-cycle femtosecond pulse generation. Differences between this spectral broadening by the N_2 molecular alignment and the traditional method of the third-order nonlinear polarization of noble gases[25,26] are also discussed.

2. Theoretical simulation

2.1. Theoretical model

In this study, we use the theoretical formulas and simulation method from our previous work,[10] where the aligned refractive index modulation is determined by

$$n(t) \approx 1 + 2\pi N(\alpha_\perp + \Delta\alpha\langle\cos^2\theta(t)\rangle), \quad (1)$$

where N denotes the density of molecules and $\Delta\alpha(\omega) = \alpha_\parallel(\omega) - \alpha_\perp(\omega)$. Here $\alpha_\parallel(\omega)$ and $\alpha_\perp(\omega)$ represent the parallel and the perpendicular components of the molecular polarizability, respectively. The $\langle\cos^2\theta(t)\rangle$ is a thermally averaged quantum value that characterizes the degree of alignment and is obtained by solving the Schrödinger equation

$$\mathrm{i}\frac{\partial\psi(\theta,t)}{\partial t} = \left(\frac{J^2}{2I} - \Delta\alpha\pi|E_{\mathrm{pump}}(t)|^2\cos^2\theta\right)\psi(\theta,t), \quad (2)$$

where J denotes the angular momentum operator, I represents the moment of inertia and $|E_{\mathrm{pump}}(t)|$ is the x-axis-polarized pulse envelope which aligns the molecules.

For a refractive index $n(t)$ close to unity and neglecting the back reflection, the propagation equation for an x-axis-polarized and z-axis-propagating pulse in a hollow fiber can be written as

$$\frac{\partial E(z,t)}{\partial z} + \frac{1}{c}\frac{\partial E(z,t)}{\partial t} = -\frac{1}{2c}\frac{\partial P(z,t)}{\partial t}, \quad (3)$$

where c represents the velocity of light in a vacuum.

The effect of molecular ionization is neglected when the aligning pulse is restricted to be shorter than 100 fs and weaker than 3×10^{13} W·cm^{-2}.[11] For the pulse propagation in an N_2-filled hollow fiber, we need to consider the effects of the third-order nonlinear polarization, dispersion and molecular alignment. For noble gas Ar, there is no molecular alignment.

2.2. Influence of the pulse duration

A theoretical result of the relationship between the laser intensity and the induced alignment for the pulse duration of 25 fs and intensity of 1×10^{13} W·cm^{-2} is shown in Fig. 1. The maximum alignment occurs about 85 fs after the peak intensity. This means that, in contrast to the third-order nonlinear polarization, the rotational response of N_2 has a time-delayed characteristic and its induced increase in the refractive index temporally overlaps with the pulse tail. As the frequency tuning can be estimated from $\Delta\omega = -\mathrm{d}\Delta\varphi/\mathrm{d}t$, this increasing index modulation will introduce red tunability to the aligning pulse,[10] especially to its falling edge. Therefore, compared with the noble gases, this new red-tuning process will contribute to the spectral broadening of the aligning femtosecond pulses when they propagate through molecules that can be aligned, such as N_2 molecules. This trend is consistent with the previous reports,[8,17] where the real-time influence of molecular alignment on the aligning pulse itself was discussed but with the propagation distance of only several centimeters.

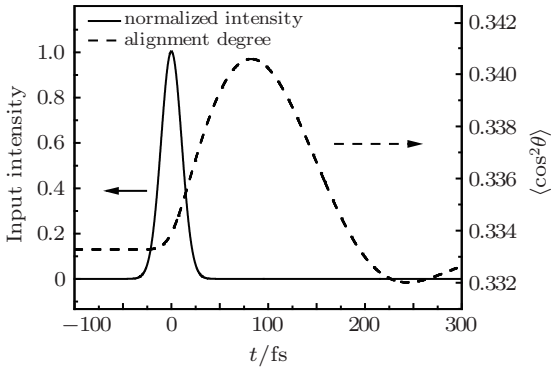


Fig. 1. N_2 alignment shown together with the aligning laser pulse. Parameters used in the simulation are 25 fs, 1×10^{13} W·cm^{-2} and 1 atm at room temperature.

To analyze the influence of this time-delayed rotational effect of alignment, we simulate the propagation of femtosecond laser pulses in a 1-m-long hollow fiber that is filled with either 1 atm Ar or N_2 at room temperature. Intensities for both input Fourier-transform-limited (FTL) pulses are 1×10^{13} W·cm^{-2}; the duration is 25 fs in Fig. 2(a) and 100 fs in Fig. 2(b). Figure 1 indicates that the rotational alignment is effective over a duration of about 85 fs. The greater the overlap between this rotational index modulation and the pulse tail, the larger the spectral red-shift of the pulse. The tail of the 25-fs laser pulse only occupies the first quarter of this increasing index modulation. Thus, compared with the third-order nonlinear polarization, the time-delayed rotational effect has little influence on the spectral broadening. This is why the spectral difference is very small between Ar and N_2, as shown in Fig. 2(a). In contrast, a 100-fs pulse basically takes full advantage of the time-delayed rotational modulation and its red tuning becomes a major contribution to the spectral broadening. This makes the spectral difference between Ar and N_2 considerable, as is evident in Fig. 2(b), especially on the red region of the spectrum. Therefore, the time-delayed molecular alignment induces an asymmetric spectral modulation of the femtosecond laser pulse, with more broadening observed on the red rather than the blue end of the spectrum. Compared with the pulse with a duration of 25 fs, this effect is more obvious for the 100-fs pulse. This result is consistent with the previous report.[17]

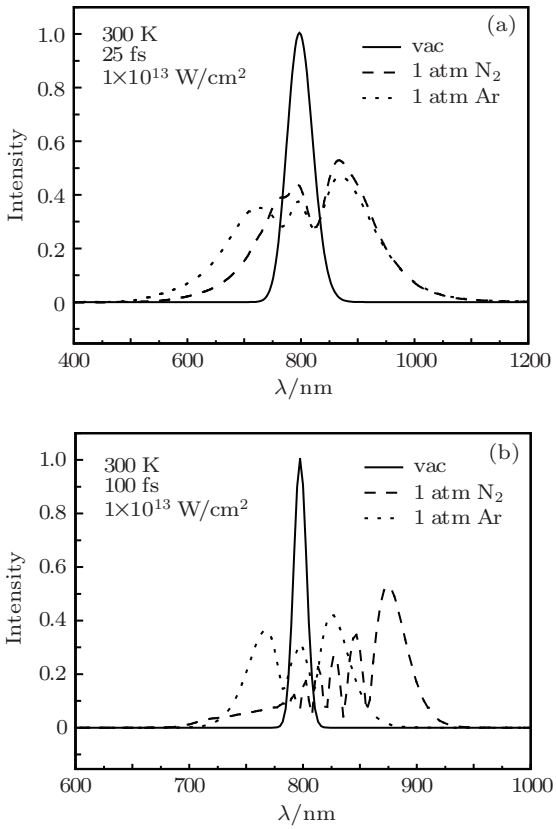


Fig. 2. Simulation results at input (solid line) and output of the N_2-filled (dashed line) or Ar-filled (dotted line) hollow fiber using an initial pulse duration of (a) 25 fs and (b) 100 fs.

2.3. Influence of the gas temperature

It has been reported that the rotationally excited effect of molecular alignment is stronger with a higher pulse intensity[27] and a lower gas temperature.[23,28,29] As the frequency tuning can be estimated from $\Delta\omega = -\mathrm{d}\Delta\varphi/\mathrm{d}t$, it is the rate of the alignment-induced index modulation that controls the frequency tuning. A detailed theoretical description of how the gas temperature influences the degree and rate of the initial alignment is provided in Fig. 3. As has been reported previously,[23,24] both the degree and rate of alignment are enhanced appreciably when the gas temperature decreases. However, as is evident in Fig. 3, this low temperature enhancement for both degree and rate of alignment is not uniform for all pulses of different durations, it is more obvious for the 100-fs pulse compared with the 25-fs pulse.

Considering the results presented in Figs. 2 and 3, we can see that the longer pulse duration of 100 fs has advantages for the alignment-induced spectral broadening for two reasons. First, the time-delayed index modulation is better exploited. Second, the enhancement of the alignment rate is more effective at low temperature. Spectral results for a 100-fs laser pulse propagating through an N_2-filled hollow fiber are presented in Fig. 4, where different lines represent spectra measured at different gas temperatures. The two panels in Fig. 4 have different input chirp signals. Figure 4(a) shows an FTL 100-fs pulse, whereas Fig. 4(b) depicts a negatively chirped pulse from an FTL 20-fs pulse. It is obvious that in both situations, the output spectrum at 90 K is more

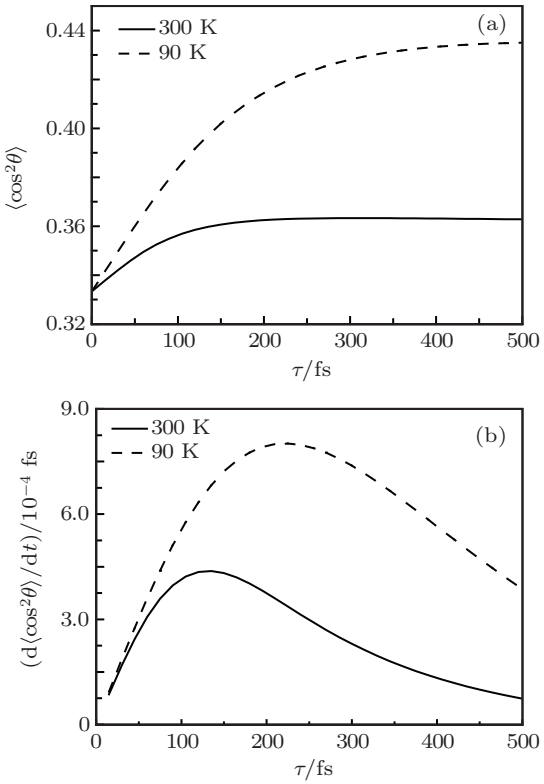


Fig. 3. (a) Maximum degree and (b) rate of the initial alignment versus pulse duration. All the peak intensities are 1.0×10^{13} W·cm^{-2}.

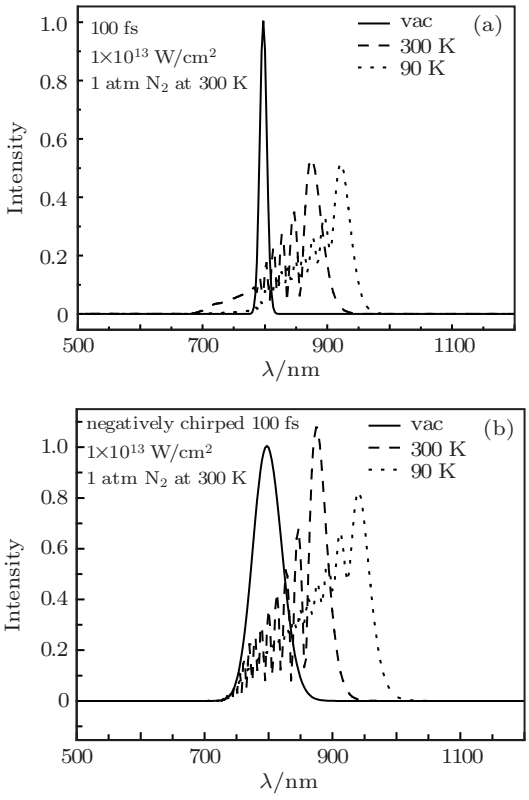


Fig. 4. Simulation results at different gas temperatures. (a) FTL 100-fs pulse and (b) negatively chirped 100-fs pulse from an FTL 25-fs pulse are shown. The N_2 molecular density is 2.68×10^{25} m^{-3}, which corresponds to 1 atm at 300 K.

asymmetric than that at 300 K, with a red-shift of about 50 nm for both peaks. Furthermore, the full width at half maximum (FWHM) of the output spectrum at 90 K is about 100 nm in Fig. 4(b), which corresponds to an 8-fs FTL pulse.

2.4. Theoretical conclusion

In conclusion, the effect of molecular alignment differs from the traditional femtosecond pulse spectral broadening by the third-order nonlinear polarization of noble gases in three ways. First, it is suitable for the longer 100-fs pulse; second, because of the low temperature enhancement of the alignment rate, stronger spectral broadening can be obtained at a lower temperature; third, due to its asymmetric broadening characteristics, the central wavelength is tuned to longer wavelengths.

3. Experiment

3.1. Experimental setup

The experimental setup is presented in Fig. 5. The beam splitter (BS) divided the output laser pulse into two parts, we used the weaker transmitted part to achieve real-time pulse duration monitoring with an autocorrelation analyzer, and the reflected part, which possessed the major output energy, was propagating through a hollow fiber. The diameter of the hollow fiber was 260 μm. The laser repetition rate was 3 kHz and a chirp modulator was used to adjust the pulse duration from FTL 25 fs to hundreds of femtoseconds. By using a positive lens, the output pulse was coupled into the 1-m-long gas-filled hollow fiber positioned in a vacuum chamber. The hollow fiber was filled with either Ar or N_2 depending on the required experimental condition. At the output side, spectral results were collected with a spectrometer.

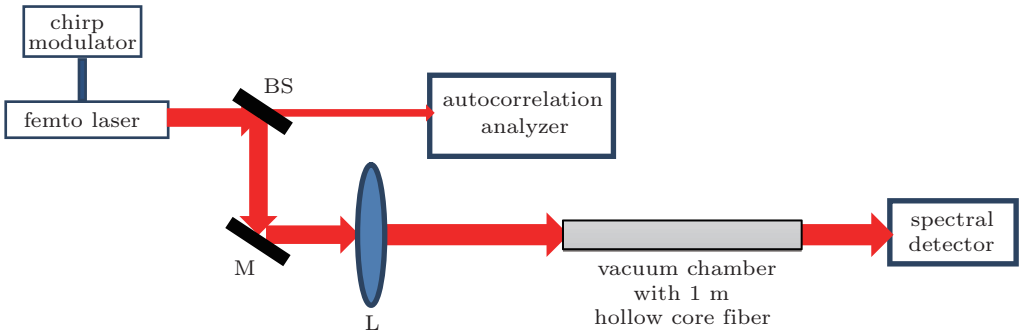


Fig. 5. (color online) Schematic representation of the experimental setup. Here BS is a beam splitter, M is a total reflector and L represents the convergent lens for laser-fiber coupling with a focal length of 1 m.

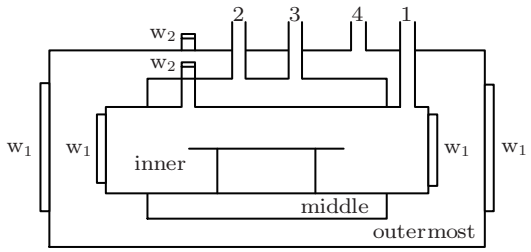


Fig. 6. Columnar vacuum chamber for experiments at different temperatures. The three layers are represented as inner, middle and outermost. Port 1 is the input and output port of the gas. The entrance and exit of liquid oxygen are through ports 2 and 3, respectively. Port 4 is the exit port of the outermost vacuum layer. The w_1 represents an optical window for laser input and output; w_2 represents an window for observing the laser-fiber coupling.

To perform experiments at different temperatures, our columnar vacuum chamber had three layers, as shown in Fig. 6. The inner layer was filled with the experimental gas (Ar or N_2) and contained an optical window on each side. The hollow fiber was also placed inside this layer. The middle layer was located around the inner while keeping the two optical windows clear. This layer could be filled with liquid oxygen for cooling. Here we did not use liquid nitrogen for cooling because liquid nitrogen would liquefy the N_2 gas in the inner layer. The outermost layer was a vacuum to guarantee the cooling process. During experiments at both room and low temperatures, the vacuum of the outermost layer was maintained. When the middle layer was filled with liquid oxygen, the N_2 gas in the inner layer could be cooled to 90 K.

We analyzed the central wavelengths at both input and output to study the characteristics of the alignment-induced spectral broadening. We based the weighted central wavelength and processing method on the concept of the weighted central frequency

$$\omega_0 = \frac{\int |I(\omega)|^2 \omega \, \mathrm{d}\omega}{\int |I(\omega)|^2 \, \mathrm{d}\omega}. \tag{4}$$

For the central wavelength, this formula yields

$$\lambda_0 = \frac{\int |I(\lambda)|^2 \, \mathrm{d}\lambda}{\int (|I(\lambda)|^2/\lambda) \, \mathrm{d}\lambda}. \tag{5}$$

3.2. Results and discussion

As discussed in Section 2, the low temperature enhancement of the alignment rate induces larger spectral broadenings

at low temperature. Experimental results obtained at different temperatures for the 25-fs input pulse are presented in Fig. 7. The spectrum at low temperature does exhibit broadenings on the red end of the spectrum; this information is presented in Table 1. It is found that the red-shift of the central wavelength at 90 K is 8 nm larger than that at room temperature. However, the spectral differences at different temperatures are small because of the small enhancement at low temperature when the pulse duration is 25 fs (see Fig. 3(b)).

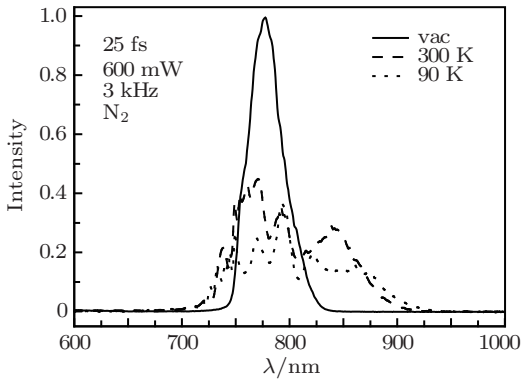


Fig. 7. Input spectrum (solid line) shown together with those after propagating through the N_2-filled hollow fiber at room temperature (dashed line) and 90 K (dotted line). The molecular density is 2.69×10^{25} m^{-3}, which corresponds to 1 atm at room temperature.

Table 1. Central wavelengths at both input and output using a 25-fs pulse.

	Input	300 K	90 K
Central wavelength/nm	776	790	798

To take advantage of the time-delayed rotational effect and its low temperature enhancement, experiments involving the longer 100-fs input pulse were also performed. Specifically, a chirp modulator was used to stretch the original FTL 25-fs pulse and its duration was measured simultaneously with an autocorrelation analyzer. In the chirped pulse situation, a higher power of 1100 mW could be used to keep a good output mode. The peak intensity at the power of 1100 mW with the chirped 100-fs pulse corresponded to that using a power of less than 300 mW when the pulse duration was 25 fs. The peak intensity was shown to be relatively low in our experiment, where the effect of the third-order nonlinear polarization was very weak and the effect of molecular alignment mainly contributed to the spectral modulation. Additionally, considering the bearing capacity of the chamber window and the need for the ground-coupled laser mode, the maximum gas pressure we used was 1.5 atm.

Results obtained at different temperatures with the longer 100-fs input pulse are provided in Fig. 8. Spectral broadenings at both gas pressures were enhanced as the gas temperature decreased. The output spectrum with 1.5 atm N_2 was broadened to an FWHM of about 80 nm at 90 K (the dotted line in Fig. 8(b)), corresponding to an FTL pulse of about 10 fs. This spectral broadening is about twice that observed at room temperature (the dashed line in Fig. 8(b)). In addition, as shown in Table 2, the spectral asymmetry became obvious. The maximum red-shift of the central wavelength was 68 nm at 90 K, but was only 30 nm at 300 K.

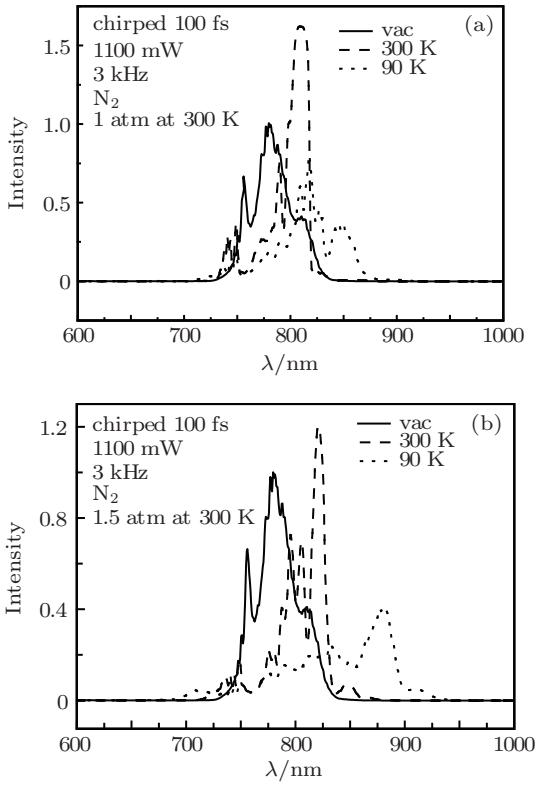


Fig. 8. Input spectrum (solid line) shown together with spectra observed after the pulse propagates through the N_2-filled hollow fiber at room temperature (dashed line) and 90 K (dotted line). The molecular density is (a) 2.69×10^{25} m^{-3} that corresponds to 1 atm at 300 K and (b) 4.03×10^{25} m^{-3}, which corresponds to 1.5 atm at 300 K.

Table 2. Central wavelengths at both input and output using a 100-fs pulse.

	Input	300 K	90 K
Central wavelength in Fig. 8(a)/nm	782	805	818
Central wavelength in Fig. 8(b)/nm	782	812	850

This is the first experimental evidence that the rate of molecular alignment is enhanced at low temperature and the enhancement degree depends on the pulse duration. The almost 200% enhancement for both spectral broadening and red shift of the 100-fs laser pulse indicates that a shorter few-cycle femtosecond pulse with a longer central wavelength can be obtained at low temperature.

4. Conclusion

The low temperature enhancement of the alignment-induced spectral broadening and the red-shift of femtosecond laser pulses was studied theoretically and experimentally. The enhancement was obvious for the 100-fs pulse, but weak when the pulse duration was 25 fs. When the 100-fs pulse, which

was negatively chirped from the Fourier-transform-limited 25-fs pulse, propagated through a 1-m-long N_2-filled hollow fiber at 90 K, its central wavelength was tuned about 68 nm towards the red end of the spectrum and the output spectrum was broadened sufficiently to generate a 10-fs few-cycle laser pulse.

References

[1] Lin G H, Heritage J P, Gustafson T K, Ghiao R Y and McTague J P 1976 *Phys. Rev. A* **13** 813
[2] Ripoche J, Grillon G, Prade B, France M, Nibbering E, Lange R and Mysyrowicz A 1997 *Opt. Commun.* **135** 310
[3] Friedrich B and Herschbach D 1995 *Phys. Rev. Lett.* **74** 4623
[4] Seideman T 1995 *J. Chem. Phys.* **103** 7887
[5] Rosca-Pruna F and Vrakking M J J 2001 *Phys. Rev. Lett.* **87** 153902
[6] Loriot V, Tzallas P, Benis E P, Hertz E, Lavorel B, Charalambidis D and Faucher O 2007 *J. Phys. B* **40** 2503
[7] Rouzée A, Guérin S, Faucher O and Lavorel B 2008 *Phys. Rev. A* **77** 043412
[8] Nibbering E T J, Grillon G, Franco M A, Prade B S and Mysyrowicz A 1997 *J. Opt. Soc. Am. B* **14** 650
[9] Bartels R A, Weinacht T C, Wagner N, Baertschy M, Greene C H, Murnane M M and Kapteyn H C 2001 *Phys. Rev. Lett.* **88** 013903
[10] Zhong F, Jiang H and Gong Q 2009 *Opt. Express* **17** 1472
[11] Wang F, Jiang H and Gong Q 2012 *J. Opt. Soc. Am. B* **29** 232
[12] Wu J, Cai H, Peng Y and Zeng H 2009 *Phys. Rev. A* **79** 041404
[13] Wu J, Cai H, Couairon A and Zeng H 2009 *Phys. Rev. A* **79** 063812
[14] Wu J, Cai H, Couairon A and Zeng H 2009 *Phys. Rev. A* **80** 013828
[15] Wu J, Cai H, Peng Y and Zeng H 2009 *Opt. Express* **17** 5822
[16] Wu J, Cai H, Zeng H and Couairon A 2008 *Opt. Lett.* **33** 2593
[17] Nisoli M, Stagira S, Silvestri S De, Svelto O, Sartania S, Cheng Z, Lenzner M, Spielmann Ch and Krausz F 1997 *Appl. Phys. B* **65** 189
[18] Litvinyuk IV, Lee K F, Dooley P W, Rayner D M, Villeneuve D M and Corkum P B 2003 *Phys. Rev. Lett.* **90** 233003
[19] Li Y, Yang S P, Jia X Y and Chen J 2010 *Chin. Phys. B* **19** 043303
[20] Velotta R, Hay N, Mason M B, Castillejo M and Marangos J P 2001 *Phys. Rev. Lett.* **87** 183901
[21] Tong A H, Liao Q, Zhou Y M and Lu P X 2011 *Acta Phys. Sin.* **60** 043301 (in Chinese)
[22] Wang Y Q, Chen D Y, Xia Y Q, Fan R W and Lu F M 2010 *Chin. Phys. B* **19** 023203
[23] Masihzadeh O, Baertschy M and Bartels R A 2006 *Opt. Express* **14** 2520
[24] Wang F, Jiang H B and Gong Q H 2012 *Chin. Phys. B* **21** 054212
[25] Hentschel M, Cheng Z, Krausz F and Spielmann C 2000 *Appl. Phys. B* **70** S161
[26] Nisoli M, Silvestri S D and Svelto O 1996 *Appl. Phys. Lett.* **68** 2793
[27] Xu N, Wu C, Huang J, Wu Z, Liang Q, Yang H and Gong Q 2006 *Opt. Express* **14** 4992
[28] Kumarappan V, Bisgaard C Z, Viftrup S S, Holmegaard L and Stapelfeldt H 2006 *J. Chem. Phys.* **125** 194309
[29] Yoshii K, Miyaji G and Miyazaki K 2009 *Opt. Lett.* **34** 1651