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Pulse reshaping in nearly resonant interaction of femtosecond pulses with dense rubidium vapor

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ABSTRACT

Propagation of intense femtosecond pulses resonant with the atomic rubidium vapor results in phenomenon known as conical emission. The origin of this phenomenon is connected with self-phase modulation in time domain accompanied with spatial self-focusing for blue-detuned pulses. When the laser central wavelength is red-detuned the self-defocusing occurs. Using frequency-resolved optical gating measurements and simple modeling of pulse propagation within the linear dispersion theory it is shown that the retrieved phase of the propagated pulse, and the associated instantaneous frequency, shows evidence of both linear dispersion and self-phase modulation. These results are consistent with the theory of the intensity dependent nonlinear refraction index in medium where linear dispersion contributes significantly to pulse reshaping.

1. Introduction

Conical emission is usually observed after propagation of intense laser beam through dense medium as a ring-patterned emission around the central laser spot in the far field. It was first observed by Grischkowsky in 1970 in experiments on self-focusing of nanosecond laser pulses in potassium vapor [1]. Subsequent experiments showed that conical emission can be induced in various media with both cw and pulsed laser sources [2–5]. When nanosecond and picosecond laser sources are used, conical emission (CE) is usually observed with laser frequency tuned in the blue wing of atomic vapor resonance frequency. The CE emission spectra appear redshifted with respect to the resonance frequency. Harter et al. [2] first realized that model consisted of self-focusing of the laser beam followed by formation of filaments, generation of Rabi sidebands and refraction of four-wave parametric amplified redshifted component at the filament boundary could account for many observed features. This model proved to be useful for explaining CE features in experiments where the laser frequency is non resonant (such as in glasses or liquids) [6]. Nevertheless, some results emerged, such as observation of CE in red-detuned conditions, that required new theoretical models. Several alternative models were proposed based on different processes. Most feasible theories included self-phase modulation [1,7], cooperative fluorescence [8,9], superfluorescence [10] and Cherenkov-like emission [11] as dominant physical processes responsible for CE. Since none of the proposed models could explain all observed CE features, most recent models are based on interplay between several different competing effects.

Several experiments with ultrashort laser sources were recently performed. Ultrabroadband CE has been observed in air from the ultraviolet to infrared region under femtosecond pumping [12–14]. Conical emission in β-barium borate after femtosecond excitation was investigated and the complete angular structure of the CE accompanying the filamentation was explained well by using the nonlinear X-wave model and Cherenkov type phase matching [15]. CE, redshifted from the potassium resonance, was reported upon blueshifted 2-ps pulsed excitation [16]. However, no emission was observed when 150-fs pulses were used. Recently, a directional fluorescence emission from sodium vapor was reported [9]. The paper showed that the CE could be caused by the cooperative fluorescence emission from coherently excited atoms. In our previous work, we obtained CE with 100-fs pulses in dense Cs vapor tuned far from the Cs atomic resonance but in the range of the cesium dimer absorption [17]. The largest cone angle was observed at about 755 nm, far away from the closest D2 cesium resonance line at 852 nm. We established that this emission originates from the nonlinear behavior of index of the refraction connected with the presence of Cs_2 X-B molecular transition with the maximum of the absorption at about 760 nm. This was further supported with a direct correlation between observed cone angle and the dimer density. We established the molecular origin of the CE with spatial self-phase modulation (SPM) as a dominant

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mechanism of the CE generation. The same mechanism was found to be responsible for CE generation after propagation of nearly resonant femtosecond pulses in rubidium vapor [18]. When the laser source was tuned in the blue wing of D2 rubidium resonance line (780 nm), self-focusing occurred together with CE. The CE spectra approximately followed the spectra of incident pulse i.e. no redshift of CE was observed. Spatial characterization of CE showed that for the far-blue-wing excitation, the CE consists of a bright ring without a central spot. The measured magnitude of the cone angle as a function of detuning from the resonance obeyed dependence of the form $\Theta_{sf} \propto \Delta^{1.5}$, Θ being cone angle and Δ laser frequency detuning, which is in accordance with the theory of self-focusing in the adiabatic following limit [1,19]. When the incident laser pulse was tuned in the red wing of both D2 and nearby D1 resonance line, self-defocusing was observed due to the negative nonlinear index of refraction. Cross-correlation measurements revealed break-up of the input pulse and extreme lengthening in time domain for both self-focused and self-defocused beams. For the laser tuned at two photon $5s_{1/2} \rightarrow 5d_{3/2}$ resonance at 778 nm, emission around 762 nm only in forward direction was observed. This is explained in the terms of resonantly enhanced four-wave mixing (FWM). These results differed significantly from previous experiments on CE induced by femtosecond, nearly resonant, blue-detuned pulses with higher energy and lower repetition rate. This confirms assumption that under particular experimental conditions one or the other mechanism is responsible for the generation of conical emission. An interesting application of switch from self-focusing to self-defocusing upon scanning through the atomic resonance has been recently used for error signal generation applied to stabilize the laser frequency [20].

In order to gain deeper insight into the propagation of fs pulses in near resonant conditions, the spectral and temporal intensity and phase of the propagated pulse are determined by Frequency-resolved optical gating (FROG) and presented in this work. The FROG using second harmonic generation (SHG) is well established technique for complete characterization of ultrashort pulses [21]. The use of second-order nonlinear process enables measurements of very weak pulses. A modification of FROG called XFROG uses cross-correlation between the transform limited pulse and the pulse which has undergone some modification due to propagation. Ambiguity in time direction, usually associated with auto-correlation FROG measurements is removed in XFROG measurements. When an ultrashort pulse experiences self-phase modulation its shape both in time and in frequency domain becomes increasingly complex. Such pulses are formed, for instance, in nearly resonant interaction of ultrashort pulses with dense atomic vapor. The motivation for using FROG in characterization of CE pulses came from the fact that knowing the pulse electric field, the information on the type of interaction experienced by the pulse propagating through the nearly resonant medium can be extracted. For example, pulses that experienced SPM usually have phase proportional to the field intensity. When instantaneous frequency of the pulse is calculated, it can be directly compared to the SPM connected with positive or negative index of refraction. Furthermore, self-steepening leading to optical shock-wave formation could be seen in measured pulse intensity in time domain.

In this paper we report FROG and XFROG measurements in rubidium vapor at several temperatures and at two different laser central wavelengths, one red-detuned and one blue-detuned from the atomic resonance. Here we show that the FROG retrieved phase and the associated instantaneous frequency of the propagated nearly resonant blue and red-detuned pulses are strongly affected by linear dispersion and nonlinear effects such as self-phase modulation. Significant discrepancy in the observed frequency shift from the theoretical model of linear propagation is in accordance with the theory of adiabatic following (AF) [1,19]. The AF theory predicts the sign change of the non-linearity between positively and negatively detuned pulses. We show that this non-linearity sign change results in reversed chirp sign compared to the change in the pulse chirp sign resulting from the linear dispersion theory.

2. Methods

The intensity cross-correlation measurements can provide only limited information on the effects of propagation through dense vapor medium on nearly resonant femtosecond pulses [18]. In order to obtain complete pulse characterization frequency-resolved optical gating was performed, which already showed to be useful for characterization of $0-\pi$ pulses produced after propagation of femtosecond pulses through resonant, low density rubidium vapor [22]. The main difference and practical challenge lies in the fact that self-focusing and defocusing demand introduction of additional optical element for re-collimation of CE beam. In our measurements a convergent lens was inserted after the focal point in order to keep the beams overlapped and focused in the nonlinear crystal. Since the focal point changed its position as the vapor density was changed, the lens was repositioned for each measurement. Lens position also depends on the tuning of the pulse central wavelength to blue or red wings of rubidium resonance lines since this determines whether the laser beam will experience self-focusing or self-defocusing, respectively.

Similar to work of Skenderović et al. [18], conical emission is generated when nearly transform-limited femtosecond pulses with duration of about 80 fs and repetition rate of 80 MHz propagate through dense rubidium vapor confined in 120 mm long cylindrical all-sapphire cell. The heaters in the oven are constructed in such a way that the windows of the cell are kept at slightly higher temperature than the body of the cell, preventing deposition of the rubidium on the windows and enabling stable long-term operation of the cell. Mode-locked Ti:sapphire laser oscillator (Spectra Physics Tsunami) delivered 10 nJ pulses with central wavelength tunable in the 740–880 nm range. In order to explore the nature of CE, the central wavelength was chosen to be either at 760 nm, blue-detuned with respect to the Rb resonance lines (780 nm and 795 nm), or red-detuned, centered at 820 nm. The pulse spectral bandwidth was 11 nm FWHM. When the laser pulse spectrum is exactly at the resonance (780 nm) the pulse is almost entirely absorbed at elevated Rb densities, making FROG measurements unfeasible. The nearly Gaussian beam with beam waist of ≈ 4 mm entered the Rb cell unfocused. For both cross-correlation and auto-correlation multiple shot measurements the temporal delay between the two correlated pulses is obtained using delay stage resulting in minimal temporal step of 2 fs. The two parallel beams were focused onto a 100 μm thick nonlinear BBO crystal using a $f=200$ mm spherical mirror. The generated SHG signal was spectrally resolved using compact CCD spectrometer (Ocean Optics HR4000) with spectral resolution of about 1 nm. The spectra were collected with integration time between 30 and 50 ms and average of 10 spectra was used in FROG analysis. The laser beam that propagated through dense rubidium vapor experiences either self-focusing or self-defocusing. Therefore, the beam was collimated after the cell using $f=700$ mm convergent lens. For lower vapor densities (vapor temperature below 330 °C) nonlinear effects become negligible and beam collimation after the cell is no longer required. On the other hand, beam collimation proved to be extremely difficult at vapor temperatures above 350 °C resulting in poor FROG traces (Fig. 1).