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Observation of the near transform-limited high-resolution tunable far-ultraviolet light*

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This paper studies the near transform-limited high-resolution tunable far-ultraviolet light which is generated by narrow-band tunable light both theoretically and experimentally. It shows the theoretical work about conversion efficiency of sum-frequency, which is restricted by phase-matching angle, walk-off angle and phase distortion and so on, and sets experiment to check this work successfully. Also the origin of chirp of dye laser is analysed.

Keywords: near transform-limited, narrow-band, high-resolution, chirp effect
PACC: 3320L, 4255, 4265

High-resolution far-ultraviolet (UV) light sources are applied largely to drive the higher atomic state in the investigation[1,2] of highly excited Rydberg states, UV four-wave mixing, UV electromagnetically induced transparency (EIT) and so on. It is so important that many methods are adopted to generate far-UV light in which the most familiar one is multi-wave mixing. According to the excitation source, they include continuous wave (CW) light and ultrashort pulse (transform-limited laser which follows the Fourier transform limit $\Delta t \times \Delta f = 0.44$, where Δt is pulse-width and Δf is transform-limited line-width) multi-wave mixing. Unfortunately, as we known, there is no near transform-limited high-resolution tunable far-UV light, which is generated by narrow-band tunable dye laser.

Noisy light can be used to probe molecular dynamics, and it offers an unique alternative to the more conventional frequency domain (narrow bandwidth) spectroscopies and ultrashort (femtosecond scale) pulse time domain spectroscopies. Like the frequency domain spectroscopies, the CW nature of noisy light allows precise measurement of transition frequencies. However, like time-resolved techniques, its ultrashort noise correlation time offers the time resolution to directly measure the time domain material response. There is a noisy light analogue for nearly every current conventional optical spectroscopy. Notable exceptions are those spectroscopies that explicitly exploit the phase-locked nature of short pulses. In short, noisy light techniques offer a third means of studying molecular line shapes which complement the more familiar CW and ultrashort pulse techniques.

In this paper we report a near transform-limited high resolution tunable far-UV light which is generated by narrow-band tunable dye laser both theoretically and experimentally. We take sum-frequency generated (SFG) light as example, which includes the nonlinear phase matching angle, the walk-off angle, and investigate its conversion efficiency. Also the frequency chirp[3−5] of the pulse is analysed.

For SFG light, our interest is conversion efficiency. Many factors including fundamental beam depletion, walk-off, diffraction, imperfect phase matching and phase distortion affect the conversion efficiency. The conditions of our work are low depletion in the nega-

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tive uniaxial crystal in the ns range (3.5 ns).

In terms of the phase matching condition, the relationship of the refractive index of the fundamental beam, secondary harmonic generation (SHG) beam and SFG beam is

$$n_{\mathrm{e}}(3\omega) = \frac{n_{\mathrm{o}}(\omega)}{3} + \frac{2n_{\mathrm{o}}(2\omega)}{3}. \tag{1}$$

Thus, we attain the phase matching angle of the SFG light

$$\sin^2\theta_{\mathrm{m}} = \frac{\dfrac{1}{(n_{\mathrm{o}}(\omega)/3 + 2n_{\mathrm{o}}(2\omega)/3)^2} - \dfrac{1}{n_{\mathrm{o}}(3\omega)^2}}{\dfrac{1}{\bar{n}_{\mathrm{e}}(3\omega)^2} - \dfrac{1}{n_{\mathrm{o}}(3\omega)^2}}, \tag{2}$$

where θ_{m} is the phase matching angle. The $n_{\mathrm{o}}(\omega)$, $n_{\mathrm{o}}(2\omega)$, $n_{\mathrm{o}}(3\omega)$ are indexes of refraction of the ordinary beams (o-ray) of the fundamental, SHG and frequency- tripling beams of the SFG process, respectively. The $\bar{n}_{\mathrm{e}}(3\omega)$ is the short axis value of the ellipsoid of the index of refraction (Fig.1(a)).

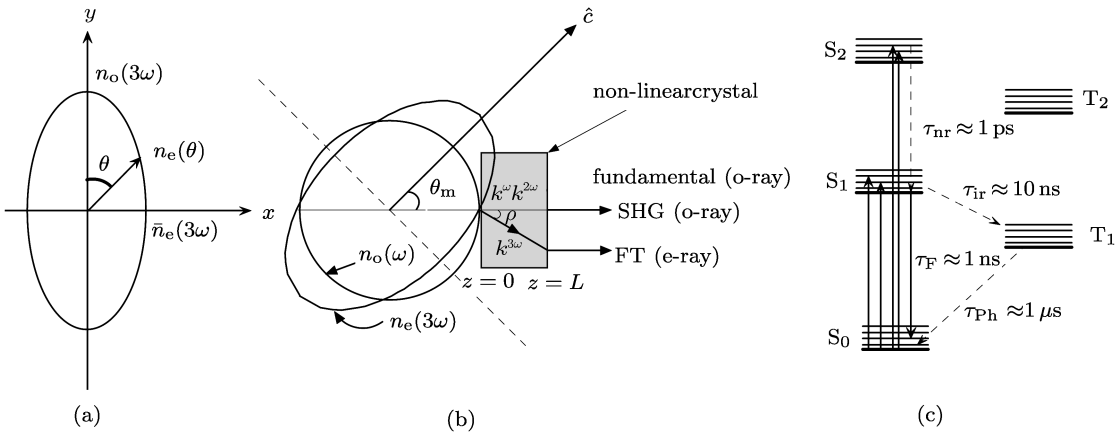


Fig.1. (a) The ellipsoid of the index of refraction of the extraordinary beam. (b) Illustrating double refraction and walk-off angle for negative uniaxial crystals. (c) Eigenstates of a typical dye molecule. The solid lines are radiative transitions, and the dashed lines are nonradiative ones.

Due to double refraction, the extraordinary beam (e-ray) moves away from the o-ray as it propagates through the crystal, limiting the interaction length for beams of a given aperture. This effect, also called the aperture effect, is another factor that limits the conversion efficiency. In terms of the definition of the walk-off angle, the walk-off angle ρ (Fig.1(b)) of the sum-frequency is determined by

$$\tan\rho = \frac{1}{2}n_{\mathrm{o}}(\omega)n_{\mathrm{o}}(2\omega)\left[\frac{1}{\bar{n}_{\mathrm{e}}(3\omega)^2} - \frac{1}{n_{\mathrm{o}}(3\omega)^2}\right]\sin(2\theta_{\mathrm{m}}). \tag{3}$$

Due to walk-off of the e-ray, the solutions of the wave equations describing the fields can no longer be found analytically because variations in the transverse directions must be taken into account. Our treatment is like that of Boyd *et al*,[6] they obtained solutions in terms of integrals which can be expressed as linear combination of error functions.

To attain higher irradiance which drives higher conversion efficiencies, the beam is often focused into the nonlinear crystal. If the confocal parameter of the diffracted beam is comparable to the length of the crystal, the plane wave approximation cannot be valid anymore, and so an even more general approach is required. Detailed model of Kleinman *et al*[7] relates the SHG field directly to the polarization of the medium (the source term) by means of a Green's function formalism. Their numerical predictions of the irradiance fit superbly with experimental data for near- and far-field limits, even when the SHG spatial profile is complicated by walk-off, diffraction and depletion. Boyd and Kleinman applied the above model to Gaussian laser beams, and solved for optimum conditions for SHG under various types of beam propagation. They listed the asymptotic scaling

behaviours of η^{SHG}.[8] We prompt their work and exploit the coupled wave equation to give the expression of the following conversion efficiency:

$$\eta^{\mathrm{FT}} = \frac{512\pi^5 d_{\mathrm{eff}}^2 I_1' I_2}{n_{\mathrm{o}}^{\omega} n_{\mathrm{o}}^{2\omega} \bar{n}_{\mathrm{e}}^{3\omega} \lambda_3^2 c(I_1' + I_2)} \cdot \begin{cases} L^2, & L_{\mathrm{a}}, L_{\mathrm{f}} \gg L, \quad (4\mathrm{a}) \\ L \cdot L_{\mathrm{a}}, & L_{\mathrm{f}} \gg L \gg L_{\mathrm{a}}, \quad (4\mathrm{b}) \\ L_{\mathrm{f}} \cdot L_{\mathrm{a}}, & L \gg L_{\mathrm{f}} \gg L_{\mathrm{a}}, \quad (4\mathrm{c}) \\ 4L_{\mathrm{f}}^2, & L \gg L_{\mathrm{a}} \gg L_{\mathrm{f}}, \quad (4\mathrm{d}) \\ {}^{19}\!/_{4} L_{\mathrm{f}}^2, & L_{\mathrm{a}} \gg L \gg L_{\mathrm{f}}, \quad (4\mathrm{e}) \end{cases}$$

where d_{eff} is the effective polarizability tensor. $\lambda_3 = 2\pi c/\omega_3$ is the vacuum wavelength of the 3ω, $L_{\mathrm{a}} = \sqrt{\pi}\omega_0/\rho$ is the aperture length defined by the walk-off angle ρ, $L_{\mathrm{f}} = \pi b/2$ is the effective length of focus, $b = 2\pi\omega_0^2/\lambda$ is the confocal parameter. ω_0 is the waist radius, I_1', I_2 are the intensity of fundamental beam and SHG beam respectively.

Compare to the femtosecond laser,[9] for which we must consider the dispersion and other effects induced by the group velocity when interactions between pulses shorter than 100 fs are considered. Equations (4a)–(4e) do not include the dispersion and pump depletion. Following a prescription by Eckardt *et al*, we will correct for this by the expression

$$\eta_{\mathrm{corrected}} = \frac{\eta^{\mathrm{FT}}}{1 + \eta^{\mathrm{FT}}}. \quad (5)$$

In addition, the phase evolution of the interacting fields is important. Phase distortions can lead to modulation of the irradiance of the fundamental field and SHG field, which in turn may limit conversion efficiency. Phase distortions are caused by a change in the refractive index of the material such as laser dye solution, crystal and so on.

The light source used by us is pulse pumped dye laser, so corruption of the optical phase during pulse amplification of a laser beam has been a dominant source of systematic uncertainty in laser spectroscopic measurements. We consider various factors that could affect the phase evolution of the pulse-amplified beam to better understand the physical origin of the phase perturbations.

Consider a monochromatic plane wave propagating in a dye solution. We ignore any variation in the transverse directions, unimportant for the geometry of the capillary cells. The instantaneous frequency is defined as

$$f_{\mathrm{inst}}(L,t) = f_{\mathrm{inst}}(0,t) - \frac{L}{2n\lambda}\frac{\mathrm{d}\chi}{\mathrm{d}t}, \quad (6)$$

where $f_{\mathrm{inst}}(0,t) = \omega_0/2\pi$ is the frequency of the field as it enters the active medium at $z = 0$, χ is the linear susceptibility of the medium and n is the refractive index of the medium. This expression describes the frequency evolution of the light traveling through any medium with a time dependent susceptibility. Now we seek an expression for $\mathrm{d}\chi/\mathrm{d}t$ appropriate for a dye solution.

In a typical dye molecule (Fig.1(c)), there is a ladder of singlet states S_0, S_1, S_2, etc, where S_0 is the ground state. There is also a ladder of triplet states T_1, T_2, etc. On absorbing energy (e.g. from a pump laser pulse), the dye molecules which are excited to a singlet state-transitions to the triplet states are spin-forbidden. The nonradiative transitions to the S_1 state from higher states are very fast (∼1 ps), so the population inversion required for amplification occurs only between S_1 and S_0. Molecules also may undergo intersystem crossing to the triplet manifold. However, such transitions occur in the time scale of a few tens to few hundreds of nanosecond, and so are not important to consider for our nanosecond laser pulses.

In a dye solution, therefore, the contributions to χ are primarily from the S_0 and S_1 states of the dye molecule, and from the solvent. We ignore higher-order susceptibilities ($\chi^{(1)}E \ll \chi^{(2)}E^2$)—in fact it will turn out that contribution from linear susceptibilities is sufficient to explain most of the phase evolution. As solvent heating is not important, we do not include its effect.

Most of the contribution to the susceptibility is predominant from the solvent. Therefore, we may replace n with the refractive index solvent of n_{s} in Eq.(6). The change in the susceptibility, however, is due only to the dye molecules in solution. The susceptibility of the solvent does not change because the frequency of the amplified light is far from any resonance in the solvent molecule, as otherwise the solvent would not be transparent. So, the susceptibility of the

dye molecules in solution is the only contributory of the changing susceptibility. Therefore, the susceptibility can be expressed as[3]

$$\chi(\omega, z, t) = \alpha_0(\omega) N_0(z, t) + \alpha_1(\omega) N_1(z, t), \quad (7)$$

where α_0 and α_1 are the real parts of the effective polarizabilities at frequency ω in the S_0 and S_1 states, respectively, and N_0, N_1 are the corresponding number densities of the dye molecules. By the 'effective', we mean that the local field corrections have been included. Therefore, α_0 is also negligible[3] and so $\chi = \alpha_1 N_1$. Equation (6) can then be expressed as

$$f_{\text{inst}}(L, t) = f_{\text{inst}}(0, t) - \frac{L}{2n_s\lambda}\alpha_1 \frac{dN_1}{dt}. \quad (8)$$

Here we have assumed that the pumping of the medium is spatially uniform and the input beam intensity is weak so the population density of S_1 does not depend on the distance traveled in the amplifier.

Equation (8) expresses the primary origin of frequency chirp and shift in dye amplification, in the small-signal limit. A pump pulse with a time-dependent irradiance causes a time-dependent excited state population. This changes the polarizability and the refractive index of the medium during the amplification process, causing a perturbation in the optical phase. Therefore, the origin of the amplifier shifts is intimately connected to the amplification process itself.

One source of index variation in the crystal is the optical Kerr effect $n = n_0 + n_2 I$, where n_0 is the linear refractive index, and n_2 is the nonlinear refraction coefficient. It is possible to relate n_2 to $\chi^{(3)}$ with which we are very familiar. In addition to a small contribution from the optical Kerr effect, we show that there is a much larger effect, originating from the $\chi^{(2)}$ susceptibility with the same general behaviour, which occurs when the phase-matching condition is not exactly satisfied. Here we give our explanation of the phase distortion due to cascaded $\chi^{(2)}$ processes attributing to self-phase modulation. The intense SFG field generated in a nonlinear medium due to $\chi^{(2)}(3\omega, 2\omega, \omega)$ interacts with the fundamental beam through the second order susceptibility $\chi^{(2)}(\omega, 3\omega, -2\omega)$ to produce another field at the fundamental frequency and $\chi^{(2)}(2\omega, 3\omega, -\omega)$ to produce another field at the SHG frequency. The generated fields then interferes with the residual fundamental beam and SHG. And if the process is not perfectly phase matched, they perturb the phase of the fundamental field and SHG field. These cascaded processes effectively mimic $\chi^{(3)}$ term, and so can be characterized by a nonlinear effective refractive index n_2^{eff} which could be ∼10 times higher than n_2. From our experimental result, the chirp which was generated by cascaded $\chi^{(2)}$ is about 10 MHz, 10% of the total chirp. So we can see that the phase distortions which are caused by a change in the refractive index of the material are very important for conversion efficiency.

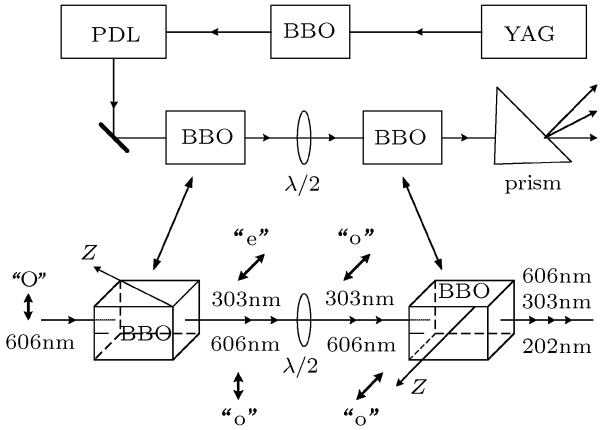


Fig.2. Experimental setup of SFG. The PDL denotes a dye laser. The BBO denotes β-BaB_2O_4 crystal.

The experimental arrangement is described as like as our previous work,[10] so only an outline is given here. As is shown in Fig.2, Nd:YAG 532 nm pulse laser pumps a near transform limited narrow-band tunable dye laser (Rg610 in Methanol; 10 Hz rate, 3.5 ns pulsewidth and 0.04 cm^{-1} linewidth). The dye laser's linewidth is about 1 GHz which is bigger only one order of magnitude than the transform-limited line-width $\Delta f = 0.44/\Delta t = 125$ MHz, where $\Delta t = 3.5$ ns is our dye laser's pulsewidth.

There are two β-BaB_2O_4 (BBO) crystals which are mounted on rotating stages. The physical dimension of BBO crystals are 7.5 mm with cut off angle 40° for the first one and 74.7° for the second one. A $\lambda/2$ wave plate for 606 nm, which changes the direction of the polarization 90° of the 606 nm beam but full-wave plate for 303 nm, is placed between the two crystals.

We obtain a SHG beam from the first BBO crystal, which is an e-ray by frequency doubling of the fundamental beam but is an o-ray for the BBO crystal and residual fundamental beam. This process is the type I phase matching condition that is o(606 nm)+o(606 nm)→e(303 nm). Then, the two beams pass the $\lambda/2$ wave plate. After that we get two beams with the same polarized direction. The second BBO crystal is mounted for SFG which also meets the type I phase matching condition o(606 nm)+o(303 nm)→e(202 nm). Because the

beams are e-rays relative to the first BBO crystal, we roll over the second crystal 90° to meet the condition (Fig.2).

We give the theoretical results in comparison with the experimental data from the handbook and show our experimental results. In terms of Sellmeier parameters for BBO by Kato,[11] we list our theoretical results by Eqs.(1)–(5) as follows: a) the phase matching angle is 74.75°; b) the sum-frequency walk-off angle is 2.79°; c) because of $L_a,\ L_f \gg L$, the conversion efficiency is $\eta^{FT} = 75.48\%$ and the correct result is $\eta = 43.01\%$. Comparing with the data measured by Eyler group,[3,4] we find that our results are perfect.

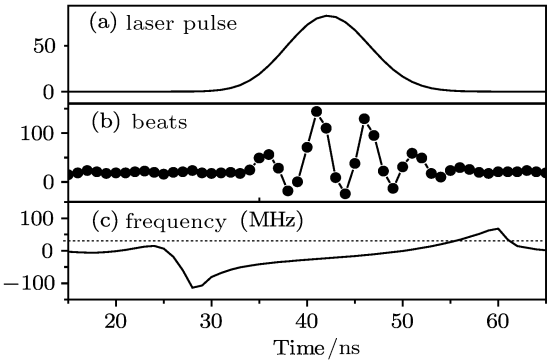


Fig.3. (a) PDA laser pulse, (b) heterodyne beats, and (c) instantaneous frequency versus time.

Using an optical heterodyne technique, we measure the perturbation of the optical phase during pulse amplification. The experimental arrangement is described in detail in Ref.[2]. The beat note (Fig.3(b)) is Fourier analysed to determine the instantaneous optical phase, which is differentiated to determine the instantaneous frequency in Fig.3(c). We have carefully determined the sign of the frequency shift, and corrected the results for a systematic underestimate of phase perturbations, revealed by numerical tests. We can see that the slop of chirp, which is caused by the polarization of dye molecule, is typically +5 MHz/ns across the laser pulse in Fig.3.

According to above theoretical results, our laboratory gets near transform-limited high resolution tunable far-UV light successfully by sum-frequency (Fig.4). In terms of our experimental setup, we get three lights in the same screen respectively. The first light is fundamental beam. The second light is SHG beam. The last small light is SFG beam. Because of the conversion efficiency, the limit wavelength got by us is 197 nm. The fundamental beam is tuned from 591 nm to 612 nm by computer in our experiment and we got tunable far-UV light from 197nm to 204 nm successfully. The SFG beam is used in our later work of the dissociation energy of H_2 experiment successfully.

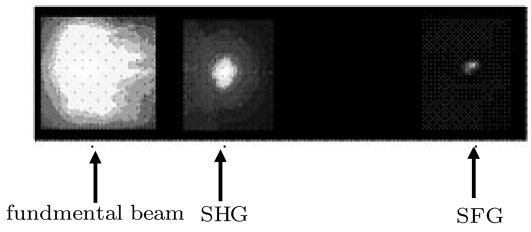


Fig.4. The typical image of near transform-limited high resolution tunable far-UV light.

In conclusion, we show our theoretical work about a near transform-limited high resolution tunable far-UV light, which is generated by narrow-band tunable light, including the nonlinear phase matching angle, the walk-off angle and the conversion efficiency, and it is compared with the experimental data which are measured by Eyler group. Then we design an experiment to check our theoretical work and get perfect results about far-UV light. In addition, we have analysed the origin of chirp effect of the dye laser and find that the origin of the amplifier phase-shifts is intimately connected to the amplification process itself.

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