Wavelength-dependence of double optical gating for attosecond pulse generation*

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Both polarization gating (PG) and double optical gating (DOG) are productive methods to generate single attosecond (as) pulses. In this paper, considering the ground-state depletion effect, we investigate the wavelength-dependence of the DOG method in order to optimize the generation of single attosecond pulses for the future application. By calculating the ionization probabilities of the leading edge of the pulse at different driving laser wavelengths, we obtain the upper limit of duration for the driving laser pulse for the DOG setup. We find that the upper limit duration increases with the increase of laser wavelength. We further describe the technical method of choosing and calculating the thickness values of optical components for the DOG setup.

Keywords: double optical gating, isolated attosecond pulse, ionization probability

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1. Introduction

In recent years, the use of high-order harmonic generation (HHG) to image and control the dynamics in atoms and molecules has received much attention.[1–3] One of the most important applications of HHG is to generate single attosecond pulses. Single isolated attosecond pulses can be used to track electron dynamics in the temporal domain, and it opens up a new field of attosecond science.[4] The single attosecond pulse has a well-defined timing and a broad continuous spectrum to access many resonant states, which have been demonstrated by the streaking experiments.[5]

Up to now, several methods have been proposed to generate single attosecond pulses. The first single attosecond pulses were generated with 7-femtosecond (fs) NIR driving pulses by selecting the XUV spectrum near the cutoff range in 2001.[6] Using shorter driving pulse, i.e., a 3.3-fs laser field, 80-as pulses centered around 80 eV have been produced and been characterized.[7] In theory, it has been shown that the two-color gating method can be an effective way to generate isolated attosecond pulses.[8,9] Using a few-cycle laser pulse in combination with a controlling field, isolated sub-100-as pulse can be produced.[10,11] Alternatively, the single attosecond pulses can also be produced by making use of the ellipticity dependence of HHG efficiency.[12,13] This method, termed polarization gating (PG), has been realized experimentally.[14] By converting a few-cycle linearly polarized driving laser field centered at 750 nm into left circularly and right circularly polarized pulses, and delaying them appropriately, the small region of temporal overlap becomes a linearly polarized field, which can produce attosecond pulses as short as 130 attoseconds.[15,16] The driving field with time-dependent ellipticity has been used in PG, in which the gate width is about half of the optical cycle.[17] In the process of generating single attosecond pulses by using the PG method, a crucial problem is the depletion of the ground-state population of the atom gas by the leading edge of circularly polarized pulses, which sets the upper limit to the pulse duration of driving laser.[18,19] In order to reduce the depletion effect, single attosecond pulse generation has been demonstrated by using a longer laser pulse, in which a weak second harmonic (SH) field is added to the driving field of PG to break the symmetry of the driving field.[20,21] This method is known as double optical gating (DOG). Compared with PG, the DOG produces the single attosecond pulses that have one cycle separation in between, instead of half cycle separation.[22,23] In both PG and DOG, the upper limit of the pulse duration can be determined, in which duration the ground state is completely depleted by the leading edge of laser field. For instance, when the intensity is 2.8×10^{14} W/cm^2 and laser wavelength is centered at 800 nm, the longest pulses for argon atom that can be used for generating single attosecond pulses with DOG, is 12 fs, and it is 6.5 fs with PG.[24]

As is well known, the cutoff of HHG is classically given as $\hbar\omega_{\rm cutoff} = 3.17U_{\rm p} + I_{\rm p}$, where $U_{\rm p}$ ($U_{\rm p} = I/4\omega^2$) is the ponderomotive energy, $I_{\rm p}$ is the ionization potential.[25,26] When the wavelength of the driving laser fields increases, the cut-

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off energy will be extended. As a result, the driving laser field with a longer wavelength has the advantage in generating more energetic photons, which will provide a potential to produce even shorter XUV pulse, i.e., zetosecond pulse. Thus, it is necessary to study the wavelength dependence of DOG to optimize single attosecond generation. In addition, to realize the experiments of PG or DOG, another important prerequisite for the implementation of them is to rightly calculate the thickness values of the optical components for PG or DOG. In this paper, we will show how to choose the thickness values of the optical components, i.e., the quartz plates and BBO, of PG and DOG setups at different driving laser wavelengths.

2. Schematic illustration of DOG

Considering the similarity between PG and DOG, we first focus our discussion on DOG in this section. The DOG method was originally realized by Mach–Zehnder interferometer method.[19] To reduce the energy loss and the temporal jitter, the interferometer method was replaced by a collinear geometry,[27] which is shown in Fig. 1.

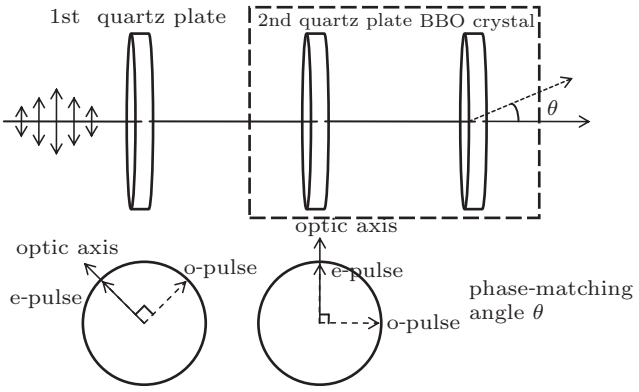


Fig. 1. Setup of collinear DOG. The linearly polarized laser pulses are sent to the first quartz plate to generate two orthogonally polarized pulses. The required group delay between them is generated by the first quartz plate, which determines the final gate width. The combination of the second quartz plate and the BBO acts as a quarter wave plate, which transforms the two linearly polarized pulses into two counter-rotating circularly polarized pulses. In PG setup, the second quartz and BBO crystals are replaced with a quarter wave plate directly. In this setup, BBO crystal is used to generate the second harmonic field.

The laser field for DOG can be given as $E(t) = E_{\rm drive}(t)i + E_{\rm gate}(t)j$,[28] where $E_{\rm drive}$ is the driving field and $E_{\rm gate}$ is the gating field of the input pulse. The driving field is given by,

$$\begin{aligned} E_{\rm drive}(t) &= E_0[(\mathrm{e}^{-2\ln 2[(t+T_{\rm d}/2)^2/\tau_\omega^2]} + \mathrm{e}^{-2\ln 2[(t-T_{\rm d}/2)^2/\tau_\omega^2]}) \\ &\quad \times \cos(\omega_0 t + \phi_{\rm CEP}) + a\mathrm{e}^{-2\ln 2(t^2/\tau_{2\omega}^2)} \\ &\quad \times \cos(2\omega_0 t + 2\phi_{\rm CEP} + \varphi_{\omega,2\omega})] \end{aligned} \quad (1)$$

and the gating field is given by,

$$\begin{aligned} E_{\rm gate}(t) &= E_0(\mathrm{e}^{-2\ln 2[(t+T_{\rm d}/2-T_0/4)^2/\tau_\omega^2]} \\ &\quad - \mathrm{e}^{-2\ln 2[(t-T_{\rm d}/2-T_0/4)^2/\tau_\omega^2]})\sin(\omega_0 t + \phi_{\rm CEP}), \end{aligned} \quad (2)$$

where E_0 is the amplitude of the circularly polarized fundamental laser field, T_0 is the laser cycle, τ_ω is the pulse duration, $\varphi_{\omega,2\omega}$ is the relative phase between fundamental and SH pulses, a represents the strength of the SH field relative to the fundamental field (for PG, a is 0), $T_{\rm d}$ is the time delay between the two circularly polarized pulses, which is given by $T_{\rm d} = L_1(1/V_{\rm ge} - 1/V_{\rm go})$, with $V_{\rm go}$ and $V_{\rm ge}$ being the group velocities of o-pulse and e-pulse in the first quartz plate respectively. Therefore, the time delay $T_{\rm d}$ is determined by the thickness of the first quartz plate L_1, which needs to introduce integer cycles of phase delay in order to keep the electric field correct and to generate two orthogonally polarized pulses. We choose two cycles of phase delay, and thus the thickness of the first quartz L_1 and the time delay $T_{\rm d}$ can be determined. The electric fields for the driving and gating fields are shown in Fig. 2. One can see that in the center of the whole pulse, the laser field is almost linearly polarized.

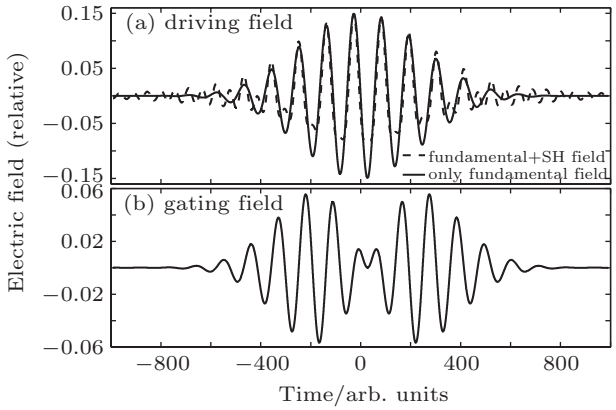


Fig. 2. Variations of driving field (a) and gating field (b) with time. In panel (a) the solid line shows the fundamental driving field without SH, i.e., the driving field of PG; and the dotted line shows the driving field of DOG, in which the SH is included. In panel (b), the gating fields are the same for DOG and PG. The electric field is nearly zero at the center of the gating field. The laser intensity is 2.8×10^{14} W/cm^2, and T_0 is 2.67 fs (corresponding to the central wavelength of 800 nm). The pulse duration is 8 fs and the ratio a of SH field to fundamental field is 0.5.

3. Discussion

In PG and DOG, the single attosecond pulse is generated by the central part of the input pulse where it is nearly linearly polarized. The HHG efficiency will increase with the decrease of the ground-state depletion effect from the leading edge of the circularly polarized pulse. Therefore, to improve the efficiency of single attosecond pulse generation, one needs to reduce the depletion effect of the leading edge and keep sufficient atoms unionized at the central part of the pulse. The depletion of the ground state as a function of input laser pulse duration can be calculated accurately by the Ammosov–Delone–Krainov (ADK) adiabatic rate.[29] Using the formula of the electric fields that have been given above, we now calculate the ground-state depletion (ionization probability) of the

leading edge of the input pulse for argon atoms through using the ADK model. The ADK formula is given by

$$W = \omega_{\rm P} |C_{n^*l^*}|^2 G_{lm} \left(\frac{4\omega_{\rm P}}{\omega_\Gamma}\right)^{2n^*-m-1} \exp\left[\frac{-4\omega_{\rm P}}{3\omega_\Gamma}\right], \quad (3)$$

$$\omega_{\rm P} = I_{\rm P}/\hbar, \quad \omega_\Gamma = E/\sqrt{2I_{\rm P}},$$

$$|C_{n^*l^*}|^2 = 2^{2^{n^*}} \left[n^*\Gamma(n^*+l^*+1)\Gamma(n^*-l^*)\right]^{-1},$$

$$n^* = (I_{\rm P}^{\rm H}/I_{\rm P})^{1/2},$$

$$G_{lm} = (2l+1)(l+|m|)!(2^{-|m|})/[|m|!(l-|m|)!].$$

Here, $I_{\rm p}$ is the ionization potential of argon atom, $I_{\rm p}^{\rm H}$ is the ionization potential of hydrogen atom, l and m are orbital angular momentum and magnetic quantum number respectively, E is the electric field amplitude, Γ is the Euler's gamma function, n^* and l^* are effective quantum numbers, and $|C_{n*l*}|$ for argon can be found in Ref. [29]. Then the probability of ionization during the time interval dt is $P(t) = \int_{-\infty}^{t} W[E(t)]{\rm d}t$. Based on the ADK model, we will study the ground-state depletion effect for both PG and DOG.

If the input pulse duration is too long, the population of the atomic ground state will be depleted by the leading edge of the input pulses, and thus PG or DOG will not work. To obtain the upper limit of input pulse duration, it is highly necessary to investigate the dependence of ionization probability on the pulse duration. As long as the atomic ground state is completely depleted, e.g., the ionization probability is very close to unit, the corresponding input laser duration is the upper limit. For different central wavelengths, the upper limit pulse duration is quite different. In Fig. 3, we show the ionization probabilities of the leading edge of the driving pulse with respect to the input pulse duration for PG. When the central wavelength increases from 800 nm to 2000 nm, clearly, the upper limit pulse duration at the same laser intensity will also increase.

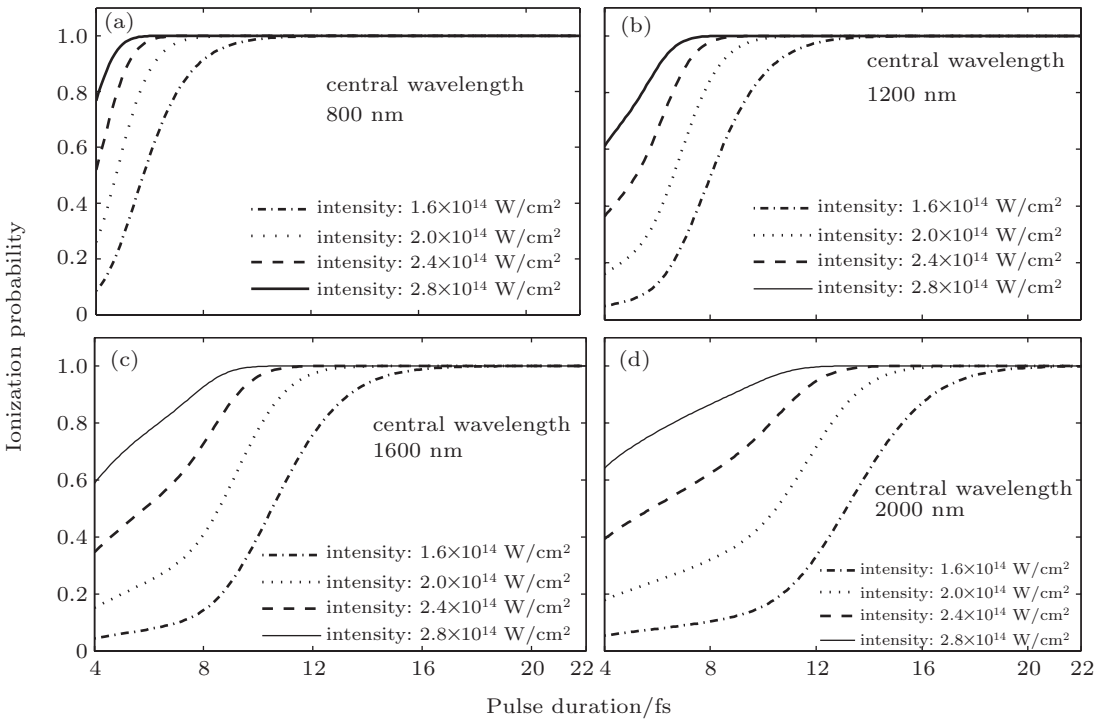


Fig. 3. Variations of ionization probability of argon atom with input pulse duration in PG for different laser wavelengths and different laser intensities. From panels (a) to (d), the central wavelengths are 800, 1200, 1600, and 2000 nm respectively. In each panel, four different curves correspond to different laser intensities from1.6×10^{14} W/cm^2 to 2.8×10^{14} W/cm^2.

In Fig. 4, we show the variations of ionization probability with input pulse duration for DOG. At the peak intensity of 2.8×10^{14} W/cm^2, the ground state will be completely depleted for the pulse duration longer than 9 fs at the wavelength of 800 nm. So the upper limit of input pulse duration is ~ 9 fs at this wavelength. The upper limit pulse durations become about 12 fs for the central wavelength of 1200 nm, 15 fs for 1600 nm, and 18 fs for 2000 nm. For the longer central wavelength, the upper limit duration becomes larger at the same intensity. Meanwhile, for each wavelength, larger laser intensity leads to a faster ground-state depletion, so that the upper limit duration becomes smaller. For instance, in Fig. 4(d), when the central wavelength is 2000 nm, the upper limit pulse duration is about 31 fs at the intensity of 1.6×10^{14} W/cm^2 and it reduces to 20 fs at the intensity of 2.4×10^{14} W/cm^2. Compared with PG, DOG can be used to generate single attosecond

pulses for longer input laser field at the same laser intensity.

Meanwhile, for isolated attosecond pulse generation with PG and DOG, the gating width is also an important parameter. The gating width is given as

$$\delta t = \frac{1}{\ln(2)} \varepsilon \frac{\tau_\omega^2}{T_\mathrm{d}},$$

where ε is the threshold ellipticity.[28] For the longer wavelength, the high harmonic yield depends sensitively on the ellipticity. To utilize the longer driving pulse, the threshold value ε will become smaller. Considering the effect of ellipticity dependence of the threshold ellipticity, the upper limit of the pulse duration can possibly be further increased.[30]

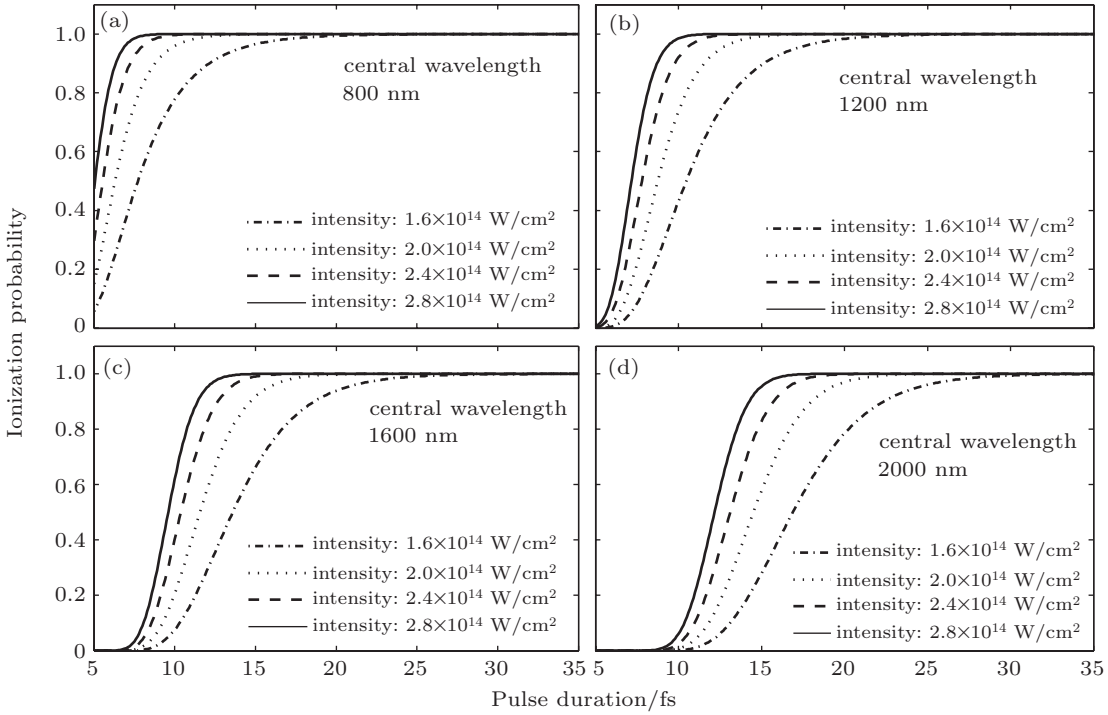


Fig. 4. Variations of ionization probability of argon atom with input pulse duration in DOG for different laser wavelengths and different laser intensities.

We have investigated the wavelength-dependent upper limit of input pulse duration for PG and DOG. When using PG and DOG, it is necessary to know how to choose the thickness values of all the optical components as shown in Fig. 1. We will describe the method of calculating the thickness values of optical components in detail. DOG is a generalized case of PG so that we only take the DOG for example. For the first quartz plate, the thickness will determine the time delay T_d. Let the refractive indexes of e-pulse and o-pulse in quartz plate be n_e and n_o and λ be the center wavelength. The first quartz plate needs to introduce a phase delay equal to an integer number of laser cycle, so its thickness L_1 should satisfy the following phase-matching equation:

$$N\lambda = L_1(n_\mathrm{e} - n_\mathrm{o}), \tag{4}$$

where N is an integer. For example, when the center wavelength is 800 nm, the thickness values of the first quartz ($n_\mathrm{e} = 1.5472$, $n_\mathrm{o} = 1.5383$) could be chosen to be 180, 270, 360, 450 μm when N is 2, 3, 4, 5 respectively.

The next point is to choose the proper thickness values of the second quartz plate L_2 and the BBO crystal L_B. The combination of these two crystal plates acts as a quarter wave plate, which turns the two orthogonally polarized pulses delayed by the first quartz plate into two counter-rotating pulses.[26] Here, two conditions must be satisfied. First, because the BBO crystal is negative uniaxial and the quartz is positive uniaxial, the phase delay of the two orthogonally polarized pulses must be matched, acting as a quarter wave plate.[31] Second, the group velocity dispersions caused by these two crystal plates should be compensated by each other. These relations are given as

$$\begin{aligned} &|[n_\mathrm{e}(\lambda) - n_\mathrm{o}(\lambda)]L_2 + [n_\mathrm{eB}(\theta,\lambda) - n_\mathrm{oB}(\lambda)]L_\mathrm{B}| \\ &= \left(n + \frac{1}{4}\right)\lambda, \end{aligned} \tag{5}$$

$$\left[\frac{1}{V_\mathrm{go}(\lambda)} - \frac{1}{V_\mathrm{ge}(\lambda)}\right]L_2 + \left[\frac{1}{V_\mathrm{goB}(\lambda)} - \frac{1}{V_\mathrm{geB}(\lambda)}\right]L_\mathrm{B} = 0, \tag{6}$$

where $n_\mathrm{oB}(\lambda)$ and $n_\mathrm{eB}(\theta,\lambda)$ are refractive indexes of o-pulse and e-pulse for the fundamental pulse in BBO with the phase-matching angle θ, respectively. The group velocities of o-pulse and e-pulse are $V_\mathrm{goB}(\lambda)$ and $V_\mathrm{geB}(\lambda)$ in BBO respectively. When we calculate the thickness values, the integer n in

Eq. (5) should be zero to reduce the error of phase match.[32] The phase-matching angle θ of type I phase-matching with BBO for each wavelength can be given by

$$\cos^2\theta = \left(\frac{\dfrac{1}{n_{\mathrm{oB}}^2} - \dfrac{1}{n_{\mathrm{eB},2\omega}^2}}{\dfrac{1}{n_{\mathrm{oB},2\omega}^2} - \dfrac{1}{n_{\mathrm{eB},2\omega}^2}} \right), \tag{7}$$

where $n_{\mathrm{eB},2\omega}$ and $n_{\mathrm{oB},2\omega}$ are the refractive indexes of e-pulse and o-pulse for second harmonic in BBO (for 800 nm, θ is about 29.2°). With solving Eqs. (5) and (6) with $n = 0$, we can obtain the thickness values of the second quartz plate L_2 and the BBO crystal L_{B}.

Because the ratio of the SH field to the fundamental field is crucial for increasing the upper limit of the input pulse duration, one should keep the conversion efficiency of the second harmonic at a high level ($> 20\%$), which depends sensitively on the laser pulse duration and the thickness of L_{B}.[33,34] The conversion efficiency of the second harmonic should be intense enough. The above calculated thickness values of L_2 and L_{B} can be well used in the DOG setup. For example, at the wavelength of 800 nm, the calculated thicknesses of L_2 and L_{B} are 450 μm and 140 μm respectively. The estimated conversion efficiency of the second harmonics is $\sim 20\%$ with this configuration.

On the other hand, if the estimated conversion efficiency is low, one should increase the thickness of BBO L_{B} to obtain a sufficient conversion efficiency. In this case, equation (6) may be not satisfied strictly. One need further consider the effect of the group velocity dispersion in Eq. (6). For instance, at a wavelength of 2000 nm, we first calculate $L_{\mathrm{B}} = 155$ μm, but the estimated conversion efficiency is about 0.08, which is too low for the DOG setup. Therefore, we increase the thickness to $L_{\mathrm{B}} = 245$ μm and the estimated conversion efficiency reaches about 0.2. And the group velocity dispersion in Eq. (6) is very small (~ 1 fs). Then one can calculate the thickness of L_2 from Eq. (5). Typically, when $N = 2$ and $n = 0$ in Eqs. (4) and (5), we can obtain the results that are listed in Table 1.

Table 1. Optimal thickness values for the optical components of DOG.

λ/μm	L_1/μm	L_2/μm	L_{B}/μm
800	180	450	140
1200	275	425	240
1600	380	315	210
2000	490	430	245

L_1: the thickness of the first quartz plate; L_2: the thickness of the second quartz plate; L_{B}: the thickness of BBO crystal.

4. Conclusion

In this paper, we systematically discussed the wavelength dependence of the DOG setup and theoretically studied the upper limit pulse durations for the driving laser wavelength ranging from 800 nm to 2000 nm. The upper limit of the driving pulse duration increases with the increase of laser wavelength and decreases with the increase of laser intensity. We also discussed how to determine the thickness values of the optical components of DOG setup in detail, i.e., the quartz plates and the BBO crystal. This study has potential to stimulate the further application of the DOG method by using the driving laser wavelength at long wavelength.

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