优化多色激光场产生单个阿秒脉冲的理论模拟

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摘要 多色组合激光场是一种产生超短孤立阿秒脉冲的可能选通方案. 以800 nm飞秒激光作为基频场, 利用遗传算法优化双色和三色激光场参数, 根据超快光场相干合成技术得到了亚周期激光场. 利用优化激光脉冲驱动氦原子即可产生超连续高次谐波并获得单个阿秒脉冲. 在考虑宏观传播效应并采取合理参数(激光强度、气体压强、气体靶长度和位置)的情况下, 利用球形空间滤波器选择合理空间范围的谐波, 得到了中心能量约为200 eV、脉宽约为50 as的单个阿秒脉冲. 理论模拟中充分考虑了目前的激光技术和实验条件, 本研究结果可为实验室产生超短阿秒脉冲提供重要的理论指导.

关键词 高次谐波, 单个阿秒脉冲, 优化激光波形, 宏观效应

阿秒(attosecond, as)脉冲是一种重要的探测和控制原子分子内部电子超快动力学行为的工具^[1~4]. 从目前技术手段来看, 阿秒脉冲主要来源于强飞秒激光与气体作用产生的高次谐波(high-order harmonic generation)^[5~7]. 从高次谐波的发射过程来看, 它实际由多个具有相同时间间隔的阿秒脉冲(即阿秒脉冲链)构成. 借助于一些“光学选通”方法^[6~8](振幅选通amplitude gating、电离选通ionization gating、极化选通polarization gating、双光学选通double optical gating等), 实验上已成功获得单个阿秒脉冲. 自从2001年成功产生单个阿秒脉冲以来^[5], 目前少数实验室已可产生50 as左右的单个阿秒脉冲^[9,10]. 产生更短阿秒脉冲仍然是一个被追求的目标.

考虑到阿秒脉冲的性能(中心能量、脉宽、强度), 从实际应用的角度来看, 高次谐波的产生过程中有两个重要的方面必须加以考虑. 第一, 谐波所能达到的最大光子能量(截止能量). 由单原子的“三步模型”可知,

对于单色驱动场, 该截止能量为 $I_p + 3.17U_p$, $U_p \propto I\lambda^2$, I 和 λ 分别为激光脉冲的峰值强度和波长. 受益于该标度关系, 利用长波中红外激光可产生能量达keV的光子. 第二, 谐波场强度. 高次谐波的转换效率较低, 对于常用的单色Ti:sapphire激光, 其转换效率为 $\sim 10^{-5}$ ^[11~13], 而对于中红外激光(2100 nm), 转换效率则低至 $\sim 10^{-9}$ ^[14]. 理论模拟结果表明, 单原子谐波场随激光波长的标度律为 $\lambda^{-5} \sim \lambda^{-8}$, 而宏观谐波场为 $\lambda^{-8} \sim \lambda^{-13}$ ^[15]. 我们之前的工作表明, 利用优化的波形则可在不降低谐波强度的前提下有效提高谐波截止能量^[16].

多色场选通(multicolor gating)是另外一种产生单个阿秒脉冲的可能方案^[6,17]. 该方法消除了驱动激光须为短脉冲(亚周期)的限制. 理论方面, 人们已提出了利用各种多色激光场驱动原子产生单个阿秒脉冲的方案^[18~28]. 在不考虑脉冲强度的情况下, 阿秒脉冲的宽度可短至10 as以下^[25,28].

需要指出的是, 在以上已有的理论模拟中只考虑

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了单原子在激光场的响应,而实验室产生高次谐波是一个宏观过程,最终收集的信号是气体靶中不同位置处的大量原子产生谐波的相干叠加结果.此时,驱动激光和各个原子产生的谐波场都在气体靶中演化,多种效应都会对总的谐波(因此也会对阿秒脉冲)产生重要影响^[29-31].

本研究开展了一个数值实验,在充分考虑真实的实验条件下,模拟利用多色激光场产生超短单个阿秒脉冲的条件和可能性.为了保证阿秒脉冲的宽度,采用遗传算法优化双色、三色激光场波形,以此来产生高次谐波.在合理的参数(激光强度、气体压强、气体靶长度和位置等)下,得到了约50 as的单个脉冲.该结果可为实验提供重要的理论指导.

1 理论方法

下面简要介绍用于计算高次谐波和阿秒脉冲的基本方法.

1.1 单原子响应

为提高计算效率,采用强场近似(strong field approximation, SFA)理论^[32]计算高次谐波.对于给定激光场,原子产生的偶极矩为

$$x(t) = i \int_{-\infty}^{\infty} d\tau \left(\frac{\pi}{\varepsilon + i\tau/2} \right)^{3/2} d^*[p_{st}(t, \tau) + A(t)] \\ a^*(t) e^{-iS_{st}(t, \tau)} \times d[p_{st}(t, \tau) + A(t - \tau)] \\ E(t - \tau) a(t - \tau) + c.c., \quad (1)$$

其中, $E(t)$ 为驱动激光场的电场, $A(t)$ 为矢势,

$a(t) = \exp\left[-\frac{1}{2} \int_{-\infty}^t w(\tau) d\tau\right]$ 为原子的基态振幅,电离速率 $w(\tau)$ 可由ADK模型^[33]计算得到, $d(t)$ 为原子从基态到连续态的偶极跃迁矩阵元.正则动量 p_{st} 和半经典作用量 S_{st} 可由式(2)和(3)计算得到:

$$p_{st}(t, \tau) = -\frac{1}{\tau} \int_{t-\tau}^t dt'' A(t''), \quad (2)$$

$$S_{st}(t, \tau) = \int_{t-\tau}^t dt'' \left\{ \frac{1}{2} [p_{st} + A(t'')]^2 + I_p \right\}, \quad (3)$$

其中, I_p 为原子的电离势.通过对 $x(t)$ 作傅里叶变换,得到单原子高次谐波谱 $S(\omega) \propto \omega^4 |x(\omega)|^2$.叠加某一能量范围的谐波,则得到阿秒脉冲.

在本研究模拟中,驱动激光场的电场可表示为

$$E(t) = \sum_{i=1}^n E_i f(t - \tau_i) \cos[\omega_i(t - \tau_i) + \phi_i], \quad (4)$$

其中, E_i 为电场振幅, $f(t)$ 为高斯脉冲包络, ω_i 为激光角频率, ϕ_i 为载波包络相位, τ_i 为相对时间延迟.参数 E_i , ω_i , ϕ_i 和 τ_i 通过遗传算法^[34]优化特定目标函数得到.遗传算法是一种借鉴生物界的进化规律而来的随机搜索算法,已广泛用于最优化问题.该算法的基本思想是:随机产生一组初始种群(个体),对它们进行交叉、变异等操作后产生新的后代,不符合条件的后代则被淘汰.经过若干代遗传后,该算法最终收敛于条件最好的染色体,得到最优解.优化中迭代代数取为3000,每代的个体数为10.采用的目标函数为

$$F = I_1 / I_2, \quad (5)$$

其中, I_1 为所需阿秒脉冲强度, I_2 为其余阿秒脉冲强度.

1.2 宏观高次谐波和阿秒脉冲

通过驱动激光场的麦克斯韦传播方程^[35]:

$$\nabla^2 E_1(r, z, t) - \frac{1}{c^2} \frac{\partial^2 E_1(r, z, t)}{\partial t^2} \\ = \mu_0 \frac{\partial J_{abs}(r, z, t)}{\partial t} + \frac{\omega_p^2}{c^2} (1 - \eta_{eff}^2) E_1(r, z, t), \quad (6)$$

得到气体靶中各点处的电场 $E_1(r, z, t)$, 式中 η_{eff} 为气体有效折射率:

$$\eta_{eff}(r, z, t) = \eta_0(r, z, t) + \eta_2 I(r, z, t) - \frac{\omega_p^2(r, z, t)}{2\omega_0^2}, \quad (7)$$

其中, 线性项 $\eta_0 = 1 + \delta_1 - i\beta_1$ 考虑了折射(δ_1)和中性原子吸收(β_1), 第二项描述了依赖于瞬时激光强度 $I(t)$ 的光学克尔非线性效应, 第三项则考虑了等离子体效应, $\omega_p(t) = [e^2 n_e(t) / (\varepsilon_0 m_e)]^{1/2}$, m_e 和 e 分别为电子质量和电荷, $n_e(t)$ 是自由电子密度.

谐波场 E_h 的三维传播方程为

$$\nabla^2 E_h(r, z, t) - \frac{1}{c^2} \frac{\partial^2 E_h(r, z, t)}{\partial t^2} = \mu_0 \frac{\partial^2 P(r, z, t)}{\partial t^2}, \quad (8)$$

其中, $P(r, z, t)$ 是极化项, 由式(1)和 $E_1(r, z, t)$ 计算得到.由气体喷嘴出口面(近场)的谐波传播到远场(收集处)满足Hankel变换.叠加特定空间和能量范围的谐波可得到宏观阿秒脉冲.

进一步利用小波变换对谐波进行时频分析^[36,37], 可得到高次谐波和阿秒脉冲的时间发射特性。

2 结果与讨论

考虑到~200 eV是光谱仪容易达到的能量探测范围, 且为了保证阿秒脉冲的强度, 选取基频场的波长 $\lambda_1=800$ nm, 相位 $\phi_1=0$, 各激光场脉宽均为6 fs. 优化过程中, 激光场的总强度为定值. 取 $I_{\text{总}}=1.0\times10^{15}$ W/cm²优化双色激光场波形. 优化得到的参数如表1所示. 此时, 为了得到最短阿秒脉冲, 第二束激光并未选取为基频脉冲的倍频谐波场. 图1(a)比较了单色800 nm激光场和优化所得到的双色场的电场波形. 可以看出, 通过相干合成两束激光得到了一个亚周期脉冲. 图1(b)给出了两种驱动场与Ne原子相互作用产生的高次谐波谱. 可以看出, 单色激光场产生的谐波谱由许多分立峰构成, 而相同能量的优化双色场产生了频宽为~140 eV的连续谱, 这更有利于合成孤立的阿秒脉冲. 另外, 双色场产生的谐波强度并未衰减, 这对于保证产生的阿秒脉冲的强度是非常重要的. 图1(c), (d)分别给出了两种激光场产生的最短阿秒脉冲. 分别叠加41~78阶、70~120阶谐波, 可以得到最短脉冲宽度分别约为75和55 as的脉冲. 但是, 单色激光场产生的是阿秒脉冲链. 双色激光场产生了强弱两个阿秒脉冲, 分别来源于电子短轨道和长轨道(图1(f)), 经过宏观演化, 长轨道进一步被抑制后, 则会得到完全的单个阿秒脉冲. 为了确认图1(c), (d)的结果, 对谐波作时频分析, 结果如图1(e), (f)所示. 对于单色激光场, 存在多次发射, 而且不同发射的能量差很小, 所以产生的连续谱很窄; 而对于双色场, 由于它的脉宽极短, 在很大能量范围内只存在一个发射, 因此可以得到单个阿秒脉冲, 而且相比单色场, 电子长轨道的贡献明显得到抑制. 由于最强阿秒脉冲来自 $t\approx0$ 时的发射, 因此它的强度和宽度 τ 与 λ_2 的依赖性较弱: $\Delta\lambda_2=5$ nm时, $\Delta\tau<5$ as.

为了得到更短阿秒脉冲, 提高驱动激光总强度, 进一步考虑三色组合场的情形. 表1给出了总峰值强度为 1.4×10^{15} W/cm²时优化的双色和三色激光场参数, 所得到的激光场波形如图2(a)所示. 可以看出, 三色场的能量更加集中, 峰值强度更强, 可以产生截止能量更高的谐波, 相对于双色场提高了约130 eV(图2(b)). 图2(c)给出了双色场产生的阿秒脉冲时域包络, 对平台区89~146阶谐波进行叠加, 得到了脉宽约为46 as的单个脉冲, 相比激光总强度为 1.0×10^{15} W/cm²时脉冲宽度得

表 1 激光总强度分别为 1.0×10^{15} 和 1.4×10^{15} W/cm²时优化的双色和三色激光场参数($I_0=1.0\times10^{14}$ W/cm²)

Table 1 Optimized laser parameters of two- and three-color fields with total peak intensity of 1.0×10^{15} and 1.4×10^{15} W/cm² ($I_0=1.0\times10^{14}$ W/cm²)

参数	数值		
	$I_{\text{总}}=10(I_0)$	$I_{\text{总}}=14(I_0)$	
	双色场	双色场	三色场
$I_1(I_0)$	6.43	9.61	6.95
$I_2(I_0)$	3.75	4.39	3.30
$I_3(I_0)$			3.75
$\lambda_2(\text{nm})$	519	515.7	574.2
$\lambda_3(\text{nm})$			489.7
$\varphi_2(\pi)$	0.23	1.71	1.46
$\varphi_3(\pi)$			0.19
$\tau_2(\text{fs})$	-0.04	0.42	0.49
$\tau_3(\text{fs})$			-0.19

到了缩短. 从图2(d)展示三色场产生的阿秒脉冲结果可以看出, 对平台区102~168阶谐波叠加, 得到了39 as的单个脉冲, 相比双色场阿秒脉冲宽度进一步减小. 叠加更高平台区182~281阶谐波同样可以得到39 as的单个脉冲. 从时频分析可以看出, 在三色场中, 电子长轨道对谐波的贡献进一步被抑制, 啁啾也更小.

将表1中列出的激光波形作为气体靶中心位置处的激光场, 计算宏观高次谐波谱及阿秒脉冲. 在模拟中进一步优化实验条件. 最终选取的参数如下: 气体靶长度为0.5 mm、气体压强为6 Torr(1 Torr=133.322 Pa)、气体靶放置在激光焦斑3.5 mm之后、各激光束腰为35 μm . 谐波信号收集器置于激光焦斑后530 mm处.

图3给出了谐波在空间和时间的发射特性. 以三色激光场为例, 从图3(a)可以看出, 作为阿秒脉冲源, 处于远场同一平面不同位置处(r)的谐波的发射时刻是完全不同的(主要由近场传播到远场的发散所引起), $r=0\sim1.5$ mm范围内的谐波发射时间相差2.5个基频场光学周期. 若用一圆孔收集这些谐波则得到脉宽很大的脉冲. 为解决此问题, 如果采用球面镜(图3(b)), 则能保证到达收集面上不同位置处的谐波具有相同的光程, 有效减小阿秒脉冲宽度. 图3(c), (d)给出了宏观谐波谱在近场、远场的空间分布, 可以看到, 谐波由近场传播到远场后发散角很小, 主要分布在传播轴($r=0$)附近, 这有利于收集谐波合成阿秒脉冲.

图1 (网络版彩色)激光总强度为 $1.0 \times 10^{15} \text{ W/cm}^2$ 的单色场和双色场产生的高次谐波及阿秒脉冲。(a) 单色场和优化双色场波形;(b) 800 nm单色场和优化双色场产生的氖原子(Ne)高次谐波谱;(c), (d) 单色场和优化双色场下分别叠加41~78阶、70~120阶谐波获得的阿秒脉冲;(e), (f) 单、双色场产生谐波的时频分析。 T 为基频场光学周期

Figure 1 (Color online) The high-order harmonics and attosecond pulses generated by single- and two-color fields with total peak intensity of $1.0 \times 10^{15} \text{ W/cm}^2$. (a) The waveforms for single- and optimized two-color laser fields; (b) the Ne harmonic spectra generated by 800-nm single-color and optimized two-color laser pulses; (c), (d) the attosecond pulses synthesized from harmonics H41–H78 and H70–H120, driven by single- and two-color fields, respectively; (e), (f) the time-frequency analysis of the harmonic spectra generated by single and optimized two-color field. T is the optical cycle of fundamental field

模拟的宏观谐波谱和合成的阿秒脉冲由图4给出。图4(a)是双色优化激光场产生的谐波在远场利用不同孔径球面镜收集的结果, 而图4(b)给出了叠加相应120~175阶谐波得到的阿秒脉冲。由于谐波谱连续性很好, 得到了单个阿秒脉冲。收集谐波的范围对阿秒脉冲的强度和宽度均有影响。若收集总的谐波, 得到的阿秒脉冲虽然强度大, 但脉冲宽度较大(77 as), 而收集 $r < 1 \text{ mm}$ 的谐波, 则可得到约51 as的单个脉冲。选用优化三色激光场(图4(c), (d)), 通过叠加 $r < 1.5 \text{ mm}$ 、

100~160阶谐波得到了47 as的单个脉冲, 而且相比双色场, 脉冲强度也得到了增强。

3 结论

通过理论模拟研究了如何利用简单可行的多色激光场产生高次谐波并进而获得单个阿秒脉冲。利用遗传算法优化单原子产生阿秒脉冲的强度和宽度, 得到了双色、三色激光场参数。相比于单色激光场产生的阿秒脉冲链, 利用多色场可以获得脉宽分别约为50和

图 2 (网络版彩色)激光总强度为 $1.4 \times 10^{15} \text{ W/cm}^2$ 时优化双色场和三色场产生的高次谐波及阿秒脉冲. (a) 两种激光场波形; (b) 两种激光场产生的Ne原子高次谐波谱; (c), (d) 叠加双色场和三色场产生的谐波得到的阿秒脉冲; (e), (f) 两种优化激光场产生高次谐波对应的时频分析

Figure 2 (Color online) The high-order harmonics and attosecond pulses generated by optimized two- and three-color laser fields with total peak intensity of $1.4 \times 10^{15} \text{ W/cm}^2$. (a) The waveforms of electric field for two laser pulses; (b) the Ne harmonic spectra generated by two laser pulses; (c), (d) attosecond pulses synthesized from harmonics generated by two- and three-color laser fields; (e), (f) the time-frequency analysis of the harmonics generated by two optimized laser fields

图 3 (网络版彩色)强度为 $1.4 \times 10^{15} \text{ W/cm}^2$ 的三色场产生的宏观高次谐波特性. (a), (b) 分别到达位于远场 $z=530 \text{ mm}$ 处的平面圆孔和球面上的谐波(100~160阶)的发射时间随径向距离的依赖关系; (c), (d) 谐波分别在近场和远场的空间分布

Figure 3 (Color online) The characteristics of macroscopic harmonics generated by three-color field with total peak intensity of $1.4 \times 10^{15} \text{ W/cm}^2$. (a), (b) The dependence of emission time of harmonics H100-H160 on their spatial distance on a circle and sphere at far field $z=530 \text{ mm}$, respectively; (c), (d) spatial distributions of harmonic emission in the near and far field, respectively

图4 (网络版彩色)在远场($z=530$ mm)利用球面滤波器收集到不同空间范围的谐波及合成的阿秒脉冲。(a), (b) 双色场, 叠加范围为120~175阶; (c), (d) 三色场, 叠加范围为100~160阶。激光总强度为 1.4×10^{15} W/cm², r 表示球面镜孔径

Figure 4 (Color online) The harmonic spectra and corresponding attosecond pulses collected by a spherical filter with different apertures at far field ($z=530$ mm). (a), (b) For two-color field the synthesizing bandwidth is from H120 to H175; (c), (d) for three-color field the bandwidth is from H100 to H160. The total laser intensity is 1.4×10^{15} W/cm², and r is the aperture for spherical filter

40 as、中心能量达200 eV的单个脉冲。由于采用的基频场为Ti:sapphire激光, 保证了阿秒脉冲的强度。

本文还模拟了宏观效应对阿秒脉冲产生的影响。结果表明, 利用优化的激光脉冲在考虑宏观效应的情况下, 仍然可以产生宽频带超连续谱, 并得到单个阿秒脉冲。通过进一步优化实验参数, 并利用球面镜消除谐

波在向远场传播过程中产生的光程差后, 最终获得了最短至47 as的孤立阿秒脉冲。

本研究所提方案比较简单、操作性强。借助于目前的实验技术, 如脉冲相干合成^[38,39]、光参量啁啾脉冲放大(optical parametric chirped-pulse amplification)等, 研究结果有望得到相关实验验证。

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Summary for “优化多色激光场产生单个阿秒脉冲的理论模拟”

Simulations on the single attosecond pulse generation by optimized multicolor laser field

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The attosecond (as) pulses are of great significance to observe and control the ultrafast electron dynamics in atoms, molecules and condensed matter. By utilizing some sub-cycle gating methods, i.e., amplitude gating, polarization gating, and double optical gating, isolated attosecond pulses (IAP) have been generated experimentally from high-order harmonics generation (HHG) by the interaction of intense femtosecond laser fields and gas targets. At present, about 50-as IAPs are now available only in few laboratories in the world. Thus, searching for the ways to generate more short IAPs is still desirable and important for the strong field physics and attosecond science.

The color gating is another potential technique generating ultrashort IAP, which could generate continuum high-order harmonics without using sub-cycle laser pulses. On the theoretical side, many schemes with two- and three-color combination field have been proposed to generate short IAP, and some works propose that in this way the IAP with width less than 10 as may be generated. However, these investigations are performed only on the single-atom level, and the real experimental conditions and strength of IAPs are not considered in their simulations.

In this work, we perform a numerical experiment on the generation of strong IAP from HHG driven by multicolor driving fields. Unlike the previous studies, the macroscopic effects are included in the present simulations. Considering the detection range of spectrograph, the centering energy of IAP is confined as ~ 200 eV. To obtain single attosecond pulse with short width, a genetic algorithm is used to optimize waveforms of two- and three-color excitation laser pulses, which is determined by intensity, wavelength and carrier envelope phase for each component. We choose the fundamental pulse as Ti:sapphire laser field with the wavelength of 800 nm to ensure the intensity of the generated attosecond pulse, and the second or third component is not confined as the commensurate harmonics of fundamental pulse. In order to possess an acceptable computation time, the model of strong field approximation is adopted in the simulations of HHG.

Our simulations show that by coherent synthesizing two or three multi-cycle pulses (the full width at half maximum of each laser field is 6 fs) we obtain sub-optical-cycle laser pulse, which can easily generate supercontinuum high-order harmonic spectra with wide bandwidth. Different from the single-color driving field, which produces the attosecond pulse train, the single attosecond pulses are generated from optimized laser fields. The width of IAP decreases with the increase of intensity of driving laser field. On the single atom level, our simulations show that about 50 as and 40 as IAP can be obtained by superimposing appropriate harmonics generated by optimized two- and three-color laser pulses, respectively. More importantly, after considering the propagation effects, about 50 as IAPs are obtained with spherical spatial filtering of harmonics in the far field, under reasonable conditions, i.e., the length and position of gas jet, gas pressure. With great advent of the ultrafast lasers technology, such as coherent waveform synthesizing, optical parametric amplification and optical parametric chirped-pulse amplification, this work gives helpful guidance for intense short IAP generation in the experiment.

high-order harmonics, single attosecond pulse, optimization of laser waveform, macroscopic effects

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