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Coherent control of terahertz harmonic generation by a chirped few-cycle pulse in a quantum well

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Abstract. We present results on the occurrence of ultrashort terahertz harmonic generation (THG) driven by a millimeter nonlinear chirped few-cycle laser pulse in a symmetric double quantum well. By solving the effective nonlinear Bloch equations, THG with a generic plateau and cutoff can be produced. The time–frequency characteristic of the ultrashort terahertz harmonic spectrum is analyzed in detail by means of the wavelet transform of induced dipole acceleration. Furthermore, an ultrabroad supercontinuum terahertz harmonic spectrum can be generated and an isolated ultrashort terahertz pulse can be obtained at the cutoff region by choosing the appropriate chirping rate parameters.

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1. Introduction

There has recently been a great deal of interest in the generation of terahertz (THz) pulses, which is generally based on the photoconductive effect, optical rectification, optical parametric conversion and so on [1]. Recently, by utilizing ultrashort optical coherent laser pulses, THz few-cycle pulses have been produced [2]–[5] because of their application in time-domain and time-resolved spectroscopies [6] and THz-induced extreme-nonlinear dynamics in quantum wells [7], etc. Planken et al [8, 9] have investigated THz radiation emitted by optically excited quantum beats in a coupled quantum well using phase-locked optical pulses. Krause et al [10] have presented THz emission in an asymmetric double-quantum-well structure via quantum control of wave packet motion. We have demonstrated [11] that the infrared spectra can be controlled by the linear chirp of the pulse in a biased semiconductor thin film.

It is well known that high-order harmonic generation (HHG) is a potential method to produce coherent high-frequency light [12]. Apart from the ‘three-step model’ [13, 14], one simple model is where a two-level atomic system is driven by a strong laser field whose frequency is much less than the transition frequency, in which HHG with the generic plateau and cutoff can also be produced [15]–[23]. For a semiconductor quantum well with only two electronic subbands driven by an intense field, the two-level model is likely to be valid for the description of HHG, because recent studies have shown that the continuum could not be taken into account in systems such as quantum wells [10, 24, 25]. Since the intersubband transition (IS) frequency is within the THz range, the generation of THz harmonics can be created in a two-subband quantum well with a strong driving field whose frequency is much less than the IS frequency.

For an ultrashort few-cycle laser pulse, the HHG spectrum will depend not only on the peaks of carrier waves but also on the carrier-envelope phase (CEP) [26]. Recently, Huang et al [27] have investigated the CEP-dependent effects of HHG in a strongly driven two-level atom. For instance, Carrera and Chu [28] showed that significant cutoff extension can be achieved by the optimization of the chirping rate parameters of a few-cycle driving pulse. We [29] have obtained an isolated 10 as pulse through the combination of a chirped laser and a static electric field. Until now, little work has been performed for the study of coherent control of HHG in the THz regime by a nonlinear chirped few-cycle laser pulse in a two-subband quantum well.

Motivated by this, in the present paper we investigate the generation of ultrashort THz harmonics in a two-subband symmetric double quantum well [30]–[32] driven by a millimeter few-cycle Gaussian laser pulse with chirping frequency. For this quantum well structure, Olaya-Castro et al [30] have derived the effective nonlinear Bloch equations on the two subband transitions. Later, by using the effective nonlinear Bloch equations, Paspalakis et al [31] studied the coherent control of electron dynamics in a symmetric double quantum well. In addition, we have investigated [32] the nonlinear propagation of ultrashort pulses on resonant IS transitions and found the signatures of self-induced transmission in multiple double quantum wells.

In this paper, by numerically solving the effective nonlinear Bloch equations, we find that the ultrashort THz harmonics with a general plateau and the cutoff can be generated, which can be explained by adiabatic state transitions. Detailed information about the harmonic generation will be presented with a time-frequency analysis by using the wavelet transform of
the induced dipole acceleration. In addition, by appropriately adjusting the chirping rates, the side peak energies of the temporal harmonic profile can be superimposed, i.e. an ultrabroad supercontinuum harmonic spectrum can be obtained driven by a Gaussian pulse with a proper chirping frequency.

2. The model and basic equations

We consider an n-type modulation-doped symmetric double semiconductor GaAs/AlGaAs quantum well. There are only two lower energy subbands that contribute to the system dynamics: \( n = 0 \) for the lowest subband with even parity and \( n = 1 \) for the excited subband with odd parity. The two subbands are driven by a p-polarized Gaussian pulse \( E(t) \) along the growth direction

\[
E(t) = E_0 \exp \left[ -4 \ln 2 \left( \frac{t-t_0}{\tau_p} \right)^2 \right] \cos \left[ \omega_p(t-t_0) + \phi(t) \right],
\]

where \( E_0 \) is the pulse amplitude, \( \tau_p \) is the pulse duration, \( \omega_p \) is the angular frequency, and \( t_0 \) is the center of the pulse. \( \phi(t) \) is the time-dependent carrier envelope phase (CEP) of the driving pulse.

The system dynamics is described by the following effective nonlinear Bloch equations [30]

\[
\partial_t S_1(t) = [\omega_{10} - \gamma S_3(t)] S_2(t) - \frac{S_1(t)}{T_2},
\]

\[
\partial_t S_2(t) = -[\omega_{10} - \gamma S_3(t)] S_1(t) + 2[\Omega(t) - \beta S_1(t)] S_3(t) - \frac{S_2(t)}{T_2},
\]

\[
\partial_t S_3(t) = -2[\Omega(t) - \beta S_1(t)] S_2(t) - \frac{S_3(t) - S_3(0)}{T_1}.
\]

Here, \( S_1(t) \) and \( S_2(t) \) are the mean real and imaginary parts of polarization, respectively, \( S_3(t) \) is the mean population inversion per electron (difference in occupation probability between the upper and lower subbands), and \( S_3(0) \) is the initial electron population. \( \Omega(t) = \Omega_0 \exp[ -4 \ln 2 (\frac{t-t_0}{\tau_p})^2] \cos[\omega_p(t-t_0) + \phi(t)] \) is the Rabi frequency of the incident laser field, \( \Omega_0 = -\mu E_0 / \hbar \) and \( \mu \) is the electric dipole matrix element between the two subbands. \( \omega_{10} \) is the renormalized but time-independent transition energy, \( \beta \) is the coefficient of the nonlinear term that renormalizes the applied field due to induced polarization, and \( \gamma \) is a result of the interplay between vertex and self-energy corrections to the transition energy. The expressions for the parameters \( \omega_{10}, \beta \) and \( \gamma \) are the same as given in [31, 32]. In addition, \( T_1 \) and \( T_2 \), which have been added phenomenologically to the effective nonlinear Bloch equations, describe the population decay time and the dephasing time in the quantum well structure, respectively.

In the two-subband quantum well model, the harmonic spectrum is obtained by the Fourier transformation

\[
P(\omega) = \left| \int \tilde{z}(t) \exp(i\omega t) dt \right|^2,
\]

where \( \tilde{z}(t) = \frac{\partial}{\partial t} \langle \Psi(t) | z | \Psi(t) \rangle = \frac{\partial}{\partial t} [N_s \mu S_1(t)] \) is the time-dependent mean dipole acceleration, \( N_s \) is the electron sheet density and \( \Psi(t) \) is the time-dependent wave function of the two-subband quantum well structure (figure 1).
Figure 1. Schematic picture of the model configuration, which comprises double $l = 5.5 \text{ nm}$ symmetric GaAs wells coupled via an AlGaAs barrier of width $d = 5.5 \text{ nm}$. The model has two subbands: $n = 0$ and 1.

Furthermore, we give a time–frequency analysis of terahertz harmonic generation (THG) by utilizing the wavelet transform of the induced dipole acceleration $\ddot{z}(t)$ [22, 33, 34] to investigate the detailed spectra and temporal structures of THG. Then it is taken as

$$A_{\omega}(t, \omega) = \left| \int \dot{z}(t') \sqrt{\omega} W[\omega(t - t')] dt' \right|,$$

where $W(x)$ is a windowed oscillating function, and is chosen to be the Morlet wavelet $W(x) = (\frac{1}{\tau}) e^{i x} e^{-x^2/2\tau^2}$. In the following, the effective nonlinear Bloch equations (equations (2)–(4)) of our model are solved numerically without making the rotating-wave approximation by means of a fourth-order Runge–Kutta method. The Fourier transformation is performed using the fast-Fourier-transform algorithm.

3. Terahertz harmonic generation

In this section, we will focus on the generation of ultrashort THz harmonics in a two-subband GaAs/AlGaAs symmetric double quantum well numerically. The structure consists of two GaAs symmetric square wells of $5.5 \text{ nm}$ width and $219 \text{ meV}$ height, coupled by an $\text{Al}_{0.26}\text{Ga}_{0.73}\text{As}$ barrier with width $d = 5.5 \text{ nm}$ as shown in figure 1. The dipole moment for the structure is $\mu = -5.50e \text{ nm}$. We take the electron sheet density to be $N_s = 5.0 \times 10^{10} \text{ cm}^{-2}$; then the system parameters $\omega_{10}$, $\gamma$ and $\beta$, which are dependent on the electron sheet density $N_s$, can be calculated to be $\hbar \omega_{10} = 4.7966 \text{ meV}$, $\hbar \gamma = 0.000965 \text{ meV}$ and $\hbar \beta = -0.779 \text{ meV}$.

The semiconductor quantum well used in this paper is designed with such a low electron sheet density that electron–electron interactions have a very small influence on our results. By comparing the effective nonlinear Bloch equations (equations (2)–(4)) with the two-level atomic system [35], it is seen that if we neglect the parameters of depolarization effects $\gamma$ and $\beta$ in the case of lower electron sheet densities, the symmetric double quantum well structure will evolve into the two-level atom system. Then, we adopt a semiclassical model in which the quantum well structure interacts with a classical Gaussian field $E(t)$, and the effective
Hamiltonian can be taken as

\[ H(t) = \hbar \begin{bmatrix} -\frac{\omega_{10}}{2} & \Omega(t) \\ \Omega(t) & \frac{\omega_{10}}{2} \end{bmatrix}. \]  

(7)

In the adiabatic basis, which means that the states follow the field, the diagonalized Hamiltonian is obtained by means of the unitary transformation

\[ \hat{U} = \begin{bmatrix} \cos \chi & \sin \chi \\ -\sin \chi & \cos \chi \end{bmatrix}, \]  

(8)

with \( \chi = -1/2 \arctan[2\Omega(t)/\omega_{10}] \). Then, this gives

\[ \tilde{H}(t) = \hat{U} H(t) \hat{U}^\dagger = \hbar \begin{bmatrix} \varepsilon_- & 0 \\ 0 & \varepsilon_+ \end{bmatrix}, \]  

(9)

where the field-dressed energies are given by

\[ \varepsilon_\pm = \pm \frac{1}{2} \sqrt{\omega_{10}^2 + [2\Omega(t)]^2}, \]  

(10)

corresponding to the up and down field-dressed adiabatic states.

In the following sections, we make sure that the adiabatic energies are avoided crossing, i.e. the avoided crossing of the adiabatic states are well separated. Then, in the adiabatic basis, the results discussed in this paper have been obtained from the numerical solution of the effective nonlinear Bloch equations (equations (2)–(4)) with the effects of electron–electron interactions on IS transitions for a two-subband quantum well system similar to the two-level atom system [35]. We assume that the system is initially in the lowest subband, so the initial conditions are \( S_1(0) = S_2(0) = 0 \) and \( S_3(0) = -1 \). In addition, we choose the relaxation times \( T_1 = 100 \) ps and \( T_2 = 10 \) ps as in [31, 32].

First, we consider the IS transition quantum well structure strongly driven by a Gaussian pulse with \( \hbar \Omega_0 = 45.80 \) meV for no time-dependent phase, \( \phi(t) = 0 \) (figure 2(a)). The pulse frequency is chosen as \( \hbar \omega_p = 1.0964 \) meV (i.e. 0.256 THz, 1.2 mm), and the duration of 1.5 cycles of the optical period \( \tau_p = 1.5 T_p = 1.5 \times 2\pi/\omega_p \). From the numerical solution of the effective nonlinear Bloch equations (equations (2)–(4)), we present the logarithm of the harmonic spectrum as a function of \( \omega/\omega_p \) in figure 2(c). As expected, the harmonic spectrum shows a plateau and a cutoff at the end of the plateau, which can be easily understood from the picture of the two-level model [20, 22]. At the level crossing times, there is a population transfer from the down adiabatic state to the up state. The quantum well system acquires energy from the field, then decays back to the down adiabatic state and radiates a harmonic of frequency

\[ \omega = N \omega_p = \varepsilon_+ - \varepsilon_- = \sqrt{\omega_{10}^2 + [2\Omega(t)]^2}. \]  

(11)

The difference between the two adiabatic states has a minimum of \( \omega_{10} \) and a maximum of

\[ N_{\text{max}} \omega_p = \sqrt{\omega_{10}^2 + [2\Omega_0]^2}, \]  

(12)

where \( N_{\text{max}} \) denotes the highest order of the harmonics. From figure 2(c) we can see that the cutoff energy from numerical results is about \( \omega_{\text{M}} = 83 \omega_p \), which is in accord with the analysis of equation (12). The frequency of harmonics corresponding to the cutoff is about 21.1 THz. So we believe that the generation of THz harmonics and mid/far-infrared harmonics can be produced in a quantum well driven by a proper moderate millimeter few-cycle laser pulse.
Figure 2. (a) The Gaussian field of $E(t)$ for $\phi(t) = 0$; the dashed lines denote the Gaussian envelope. (b) The energies of the field-dressed adiabatic states. (c) The THG spectra computed from (a). (d) The corresponding wavelet time-frequency profile.

Figure 2(b) presents the evolution of the adiabatic energies $\varepsilon_{\pm}$ from equation (10) as a function of the time $t/T_p$. It is seen that the adiabatic energies are dependent on $\Omega^2(t)$, which can be understood from the expression of $\varepsilon_{\pm}$ (equation (10)). Moreover, figure 2(d) shows the time–frequency profile of the THG spectra corresponding to the Gaussian pulse (figure 2(a)), by means of the wavelet transform of the induced dipole acceleration $\ddot{z}(t)$ from the numerical solutions of the effective nonlinear Bloch equations. Comparing figure 2(b) with (d), it is seen that the time–frequency spectra share the same shape with the positive branch of the adiabatic energies. The relationship between them can be easily found from equation (11), which indicates that the harmonic order $N = \omega/\omega_p$ is proportional to $\varepsilon_+$. In other words, the harmonic order $N$ would also be effectively determined by $\Omega^2(t)$.

Moreover, we find that THG spectra (figure 2(d)) exhibit several well-formed individual peak structures in the cutoff region, which correspond to the single peak harmonic shapes at the center time (i.e. $t = 0$ in figure 4(a)) and can in principle be superimposed with each other to produce a stronger radiation emission. However, Huang et al [27] enhanced the center energies and decreased the side ones by adding another weak field. In this paper, the THz harmonic spectrum is coherently controlled by utilizing the Gaussian pulse with a nonlinear chirping frequency, according to the relationship between the harmonic order $N$ and $\Omega^2(t)$.
Figure 3. (a) The Gaussian field of $E(t)$ for $\phi(t) = -\eta \tanh[(t - t_d)/\tau_c]$ with $\eta = 6.25$; the dashed lines indicate the envelope. (b) The energies of the field-dressed adiabatic states. (c) The THG spectra computed from (a). (d) The corresponding wavelet time–frequency profile.

For example, the time profile of CEP $\phi(t)$ considered here has the time-varying hyperbolic tangent form [28]

$$\phi(t) = -\eta \tanh [(t - t_d)/\tau_c],$$

(13)

where $\eta$ indicates the frequency sweeping range and $\tau_c$ indicates the steepness of the chirping function. $t_d$ is set at the middle of the sweep. The instantaneous frequency of the pulse is then

$$\omega_p(t) = \omega_p + d\phi(t)/dt = \omega_p - \frac{\eta}{\tau_c} \frac{1}{\cosh^2 \left( (t - t_d)/\tau_c \right)}.$$  

(14)

When $\eta = 0$, the CEP is 0, and the pulse is chirp free, which has been used above (figure 2(a)). Due to the recent advances in comb laser technology, it is highly likely that such a time-varying CEP will be achieved in the near future [28, 29, 36, 37]. As shown in [28], the chirped frequency has a large impact on the Gaussian pulse carrier waves. If we take the sweeping parameters as $\eta = 6.25$, the corresponding laser pulse is shown in figure 3(a). The amplitude of the Gaussian envelope pulse remains invariant to chirped frequency. However, the oscillatory periodicity of the reshaping Gaussian field is broken due to the chirping frequency. Thus, the Gaussian pulse with a broadener center carrier wave and two much smaller side carrier waves could be obtained. With this chirped driving pulse and a similar numerical simulation, we give the adiabatic energies together with the time–frequency and the THG
Figure 4. The temporal profiles of harmonics with different frequencies for (a) $\phi(t) = 0$ and (b) $\phi(t) = -6.25 \tanh[(t - t_d)/\tau_c]$. The different harmonic frequencies are given in the figures. $\omega_M = 83\omega$ is the cutoff harmonic.

spectra in figure 3(b)-(d). On account of the invariance of the Gaussian pulse envelope, the Gaussian pulse with chirping frequency (figure 3(a)) has the same peak value as the field in figure 2(a). Thus the highest order of the harmonic is unchangeable and the THG cutoff is also at $\omega_M = 83\omega_p$ (figure 3(c)). However, due to the contributions of chirping frequency, there is only one well-formed individual peak structure in a large harmonic region besides the THG cutoff, and the side peaks are clearly superimposed (figure 3(d)). Thus, an ultra-broad supercontinuum spectrum can be obtained in the quantum well system driven by a chirped few-cycle Gaussian laser pulse.

For a better insight into the effects of chirping frequency on the THG process, we present the temporal profiles of harmonics near the cutoff region for two cases: (a) $\phi(t) = 0$ and (b) $\phi(t) = -6.25 \tanh[(t - t_d)/\tau_c]$ by using the wavelet time–frequency analysis of the dipole acceleration $\ddot z(t)$ in figure 4. The time profiles of harmonics presented in figures 4(a) and (b) are from order $0.4\omega_M$ to $1.2\omega_M$. In both cases, the dipole profiles exhibit steep peaks at the time when the adiabatic energies reach the peak values, which contribute mainly to the THG in the field. For the chirp-free pulse, there are three peak harmonic profiles (one main inner peak profile and two small side ones) in the cutoff region; then an ultrashort THz pulse train can be generated. For the chirping driving pulse, there is only one peak harmonic profile at $\omega = \omega_M$, and an isolated ultrashort THz pulse would be obtained in the cutoff region. When $\omega$ increases to $\omega = 1.2\omega_M$, the number of peak profiles remains invariant in both cases, except that the peak intensity as well as the width harmonic profile becomes smaller as the harmonics approach the end of the cutoff region. As the harmonic order decreases from $\omega = \omega_M$ to $\omega = 0.4\omega_M$, due to the two possible times for the population transitions to occur between the adiabatic states of the two-level model, the peak profiles in both cases split into two with slight asymmetry. Also, some more side peaks appear in the case of the chirp-free pulse (figure 4(a)). But there are no additional peaks as $\omega$ decreases in the case of the chirped laser pulse (figure 4(b)), i.e. the side peaks have been effectively inhibited by using a few-cycle laser pulse with a proper chirping frequency.

We also investigate the generation of THz harmonics with different quantum well electron sheet densities $N_s$. From the numerical results, which are not shown here, we find that the THG with the generic plateau and cutoff can also be produced with larger $N_s$. However, with an
increase of $N_s$, the cutoff position would be shortened significantly due to the strong nonlinearity arising from the electron–electron interactions. Therefore, the two-subband symmetric double quantum well should be designed with lower electron sheet density, which would be more beneficial for generation of the THz harmonics.

4. Conclusions

In conclusion, we have investigated the generation of ultrashort THz harmonics in a two-subband symmetric double quantum well strongly driven by a millimeter few-cycle Gaussian laser pulse with chirping frequency. The results have revealed that the THz harmonic spectrum with a general plateau and cutoff at the end of the plateau has been obtained by solving the effective nonlinear Bloch equations. By using the wavelet transform of the induced dipole acceleration, we found that the time–frequency spectra are proportional to the energies of the adiabatic states. In addition, by a suitable choice of the values of chirping rates, the side peaks of the temporal harmonic profile can be superimposed, i.e. the ultrabroad supercontinuum harmonic spectrum will be generated and an isolated ultrashort THz pulse can be obtained. Therefore, this is a potential approach for the generation of ultrashort THz pulses.

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