Effects of excitation frequency on high-order terahertz sideband generation in semiconductors

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New Journal of Physics 15 (2013) 105015 (10pp)
Received 23 April 2013
Published 18 October 2013
Online at http://www.njp.org/
doi:10.1088/1367-2630/15/10/105015

Abstract. We theoretically investigate the effects of the excitation frequency on the plateau of high-order terahertz sideband generation (HSG) in semiconductors driven by intense terahertz (THz) fields. We find that the plateau of the sideband spectrum strongly depends on the detuning between the near-infrared laser field and the band gap. We use the quantum trajectory theory (three-step model) to understand the HSG. In the three-step model, an electron–hole pair is first excited by a weak laser, then driven by the strong THz field, and finally recombined to emit a photon with energy gain. When the laser is tuned below the band gap (negative detuning), the electron–hole generation is a virtual process that requires quantum tunneling to occur. When the energy gained by the electron–hole pair from the THz field is less than 3.17 times the ponderomotive energy ($U_p$), the electron and the hole can be driven to the same position and recombined without quantum tunneling, so that the HSG will have large probability amplitude. This leads to a plateau feature of the HSG spectrum with a high-frequency cutoff at about $3.17U_p$ above the band gap. Such a plateau feature is similar to the case of high-order harmonics generation in...
atoms where electrons have to overcome the binding energy to escape the atomic core. A particularly interesting excitation condition in HSG is that the laser can be tuned above the band gap (positive detuning), corresponding to the unphysical ‘negative’ binding energy in atoms for high-order harmonic generation. Now the electron–hole pair is generated by real excitation, but the recombination process can be real or virtual depending on the energy gained from the THz field, which determines the plateau feature in HSG. Both the numerical calculation and the quantum trajectory analysis reveal that for positive detuning, the HSG plateau cutoff depends on the frequency of the excitation laser. In particular, when the laser is tuned more than $3.17U_p$ above the band gap, the HSG spectrum presents no plateau feature but instead sharp peaks near the band edge and near the excitation frequency.

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1. Introduction

High-order harmonic generation (HHG) results from the interaction of an intense laser with atoms or molecules. HHG provides a mechanism for generating coherent extreme ultraviolet and x-ray attosecond pulses [1, 2]. The three-step model was established to describe the physical processes of HHG [3–5]. The strong laser field tilts the binding potential and the electron escapes from the charged core of the atom or molecule through quantum tunneling; the electron is then accelerated in the free space by the laser field. When the electron recollides with the charged core, a very energetic photon is emitted. Recently, high-order terahertz sideband generation (HSG) in semiconductors was predicted [6], which has a physical mechanism similar to HHG but occurs at a much lower frequency range. In semiconductors, an electron can be excited from the valance band to the conduction band with a hole left behind. The recollisions between energetic holes and electrons accelerated by a strong terahertz (THz) field result in HSG. HSG has been experimentally demonstrated [7, 8].

A fundamental difference between HHG in atoms and HSG in semiconductors is that the electron–hole (e–h) pairs in HSG are elementary excitations caused by near-infrared (NIR) lasers. In HHG, the electrons need to overcome the binding energy by quantum tunneling and therefore the quantum trajectories that satisfy the least action condition or the stationary phase condition have only complex solutions. In HSG, the laser frequency $\Omega$ can be tuned from below to above the semiconductor band edge $E_g$, and correspondingly, the initial energy of the e–h pairs generated by the NIR laser can be tuned from negative to positive relative to the band edge. Particularly, for e–h pairs with positive excess energy $(\Omega - E_g > 0$, corresponding to ‘negative’ binding energy of atoms in HHG), the e–h pairs can be directly created by real excitation without...
quantum tunneling, and in turn the quantum trajectories may have real solutions. On the other hand, the laser can be tuned so high above the band edge that the initial velocities of the e–h pairs are too high for the THz field to drive electrons and holes into recollision. Then recombination of e–h pairs has to occur through quantum tunneling (cf quantum tunneling in creation of e–h pairs for laser tuned below band edge). Therefore, there will also be a cutoff of HSG on the excitation laser frequency.

Such consideration motivates us to investigate the dependence of the HSG in semiconductors; in particular, its plateau features, on the frequency of the NIR laser. We calculate the HSG for various excitation laser frequencies and examine the plateau features. The HSG spectrum is explained by quantum trajectories in the three-step model. The previous studies on HSG for negative detuning \( (\Omega - E_g < 0) \) show that the maximal and minimal orders of the sideband plateau are given by the cutoff law

\[
N_{\text{max}} = (E_g - \Omega + 3.17U_p)/h\omega_0, \quad (1a)
\]

\[
N_{\text{min}} = 0, \quad (1b)
\]

where \( U_p = e^2 F^2/4 \mu \omega_0^2 \) is the ponderomotive energy, with \( F, \omega_0, e \) and \( \mu \) denoting in turn the strength and frequency of the THz field, the electron charge and the reduced mass of the e–h pair. The laser detuning \( E_g - \Omega \) plays a role similar to the binding energy in atoms for HHG \[6\]. The cutoff frequencies can be derived from a classical calculation with the assumption that the electrons tunnel to the conduction band with zero initial velocity. In this paper, we will show that for positive laser detuning \( (\Omega - E_g > 0) \), the HSG plateau cutoffs do not satisfy equations \((1a)\) and \((1b)\) and depend on the detuning.

2. Model and numerical simulation

Under an intense THz field, the kinetic energy acquired by the e–h pair can be much greater than the exciton binding energy in the semiconductor and the amplitude of the relative motion can be much greater than the exciton radius \[7\], so the essential physics of the HSG can be captured by the motion of free electrons and holes without Coulomb interaction \[7, 9\]. The optical response of semiconductors under THz field is determined by the inhomogeneous Schrödinger equation of relative motion for the e–h pair (see the appendix)

\[
i \frac{\partial}{\partial t} \Psi(\mathbf{r}, t) = [(\mathbf{p} - \mathbf{A})^2 + E_g] \Psi(\mathbf{r}, t) + \mathbf{d} \cdot \mathbf{E}(t) \delta(\mathbf{r}), \quad (2)
\]

where \( \mathbf{p} \) and \( \mathbf{d} \) are the momentum and the interband dipole matrix element, respectively, \( \mathbf{A}(t) = -\frac{\mathbf{F}}{\omega_0} \sin(\omega_0 t)\hat{e}_z \) is the vector potential of the intense THz field, and \( \mathbf{E}(t) = \mathbf{E}_{\text{NIR}} \exp(-i\Omega t) \) is the NIR laser field. Here the Rydberg unit system has been adopted and the sample volume is taken as unity. We have assumed that the electron and the hole are generated at the same position described by the 3D delta function \( \delta(\mathbf{r}) \) and the dipole matrix element \( \mathbf{d} \) is a constant independent of the momentum. The optical polarization of the e–h pair is \( \mathbf{P}(t) = -\mathbf{d}^* \psi(0, t) \). The interband polarization can be expressed as

\[
\mathbf{P}(t) = i \int \mathbf{d}^* \delta(\mathbf{r}) K(\mathbf{r}; \mathbf{r}', t - t') \mathbf{d} \cdot \mathbf{E}(t') \delta(\mathbf{r}') \, d\mathbf{r} \, d\mathbf{r}' \, dt', \quad (3)
\]
The polarization strength of the

\[ N \]

time equals zero); equation (4) can be generated \[ P_N(\Omega + N\omega_0) = \int \exp[i(\Omega + N\omega_0)t] \mathbf{P}(t) \, dt \]

where the action is

\[ S(\mathbf{p}, t, \tau) = -\int_{t-\tau}^{t} d\tau'' |\mathbf{p} - \mathbf{A}(t'')|^2 + (\Omega - E_g)\tau + N\omega_0 t. \]

Here \( \tau \) denotes the delay between the recombination and the creation of the e–h pair. The action \( S(\mathbf{p}, t, \tau) \) is a phase introduced by the motion of the e–h pair with canonical momentum \( \mathbf{p} \). Due to the inversion symmetry, only the even-order sidebands (with even \( N \)) can be generated [10].

For a strong THz field, the e–h pair performs relative motion with amplitude much greater than its wavepacket diffusion range. Therefore, the motion is well captured by a few quantum trajectories that satisfy the stationary phase conditions, as given by

\[ \partial_p S = 0 \Rightarrow \mathbf{p} \tau - \int_{t-\tau}^{t} \mathbf{A}(t') \, dt' = 0, \]

\[ \partial_t S = 0 \Rightarrow |\mathbf{p} - \mathbf{A}(t - \tau)|^2 = \Omega - E_g, \]

\[ \partial\tau S = 0 \Rightarrow |\mathbf{p} - \mathbf{A}(t)|^2 - |\mathbf{p} - \mathbf{A}(t - \tau)|^2 = N\omega_0. \]

The physical meanings of these equations are: equation (7a)—return of the electron to the position of the hole after acceleration by the THz field (the velocity integrated over time equals zero); equation (7b)—energy conservation at the excitation of the e–h pair; equation (7c)—energy conservation for the sideband generation through recombination of the e–h pair. In general, the saddle points \( (t_g, \tau_n) \) may be obtained by solving equation (7).

The laser detuning \( \Omega - E_g \) determines whether or not quantum tunneling is involved in the e–h generation and recombination. If \( \Omega < E_g \), the electron enters into the continuum via quantum tunneling assisted by the strong THz field (which, for a relatively weak THz field, is understood as multi-photon-assisted e–h pair generation). The tunneling physics results in complex solutions of \( t \) and \( \tau \), which in turn leads to reduced HSG intensity. When \( \Omega \geq E_g \), real excitation of e–h pairs is possible and so are real solutions of \( t \) and \( \tau \). Without requiring quantum tunneling through a binding energy barrier, the HSG can have large amplitudes. Now that the initial velocity depends on the laser frequency, the maximum kinetic energy that the e–h pair can acquire along a close trajectory and therefore the HSG plateau cutoff will depend on the laser detuning. Moreover, if the initial energy and hence the initial relative velocity of the e–h pair are

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too large \((\Omega - E_g > 3.17U_p)\), the THz field would not be able to drive the electron and the hole back to the same position. Then only through quantum tunneling can the e–h pair recombine, which means the saddle-point equations would not have real solutions. So the HSG amplitude will be large when the excitation energy \(0 < \Omega - E_g < 3.17U_p\) and drops rapidly outside this range—the HSG presents no plateau feature when the laser frequency is above the threshold.

Substituting equation \((7a)\) into equations \((7b)\) and \((7c)\), we obtain the saddle-point equations for \(t\) and \(\tau\)

\[
\frac{\Omega - E_g}{4U_p} = \left[ \alpha \sin\left(\frac{\omega_0 t - \omega_0 \tau}{2}\right) - \beta \cos\left(\frac{\omega_0 t - \omega_0 \tau}{2}\right) \right]^2, \tag{8a}
\]

\[
\frac{N\omega_0 + \Omega - E_g}{4U_p} = \left[ \alpha \sin\left(\frac{\omega_0 t - \omega_0 \tau}{2}\right) + \beta \cos\left(\frac{\omega_0 t - \omega_0 \tau}{2}\right) \right]^2, \tag{8b}
\]

where

\[
\alpha(\tau) = \cos\frac{\omega_0 \tau}{2} - \frac{\sin \omega_0 \tau}{\omega_0 \tau/2}, \quad \beta(\tau) = \sin\frac{\omega_0 \tau}{2}.
\]

The equations can have real solutions if \(\Omega > E_g\) and not too much above the band edge, which is not possible for HHG in atoms where the binding energy has to be positive \([11]\).

Equation \((8)\) provides some insight to understand the HSG spectrum. The e–h pair gains or loses kinetic energy to generate sidebands. The dimensionless sideband shift frequency \(N\alpha_0/U_p\) in the plateau regime is shown in figure 1 as a function of the delay time for various NIR laser detuning. In our calculation, the reduced mass of the e–h pair is chosen to be 0.076\(m_e\) as in GaAs; the photon energy of the THz field is 5 meV and its strength is 30 kV cm\(^{-1}\). Such parameters yield \(U_p = 90\) meV. As shown in figure 1, the upper and lower cutoffs of the HSG plateau depend on the detuning \(\Delta \equiv \Omega - E_g\). The saddle points \(t_n\) and \(\tau_n\) are real if the laser detuning \(\Delta\) is in such a range that the e–h pair has trajectories satisfying the classical mechanics. If the laser detuning is outside this range, \(t_n\) and \(\tau_n\) are complex numbers and the trajectories have to be assisted by quantum tunneling. To fulfill the saddle point equation \((7a)\) (the condition that the electron and the hole return to the same position), the initial velocity has to be anti-parallel or parallel to the THz electric field \(\mathbf{E}(t)\). For positive laser detuning \((\Delta > 0)\), the initial relative velocity of the e–h pair is non-zero and can be along different directions. Therefore for \(\Delta > 0\), the kinetic energy the e–h pair can acquire from the THz field is a double-valued function as shown in figure 1. Shown as gaps in figures 1(d)–(e), the saddle-point equations have no real solutions for certain ranges of delay time \(\tau\) even for laser detuning \(\Delta \in [0, 3.17U_p]\). This is because when the initial velocity of the e–h pair is large, the THz field cannot drive the electron and the hole back into the same position unless the delay time is in the proper range. For negative detuning \((\Delta < 0)\), the e–h pair is generated by quantum tunneling. The saddle-point solutions of \(\tau\) are in general complex numbers (see figures 1(f) and (g)). When the emission frequency outside the plateau region \((N\alpha_0 > 3.17U_p\) or \(N < 0)\) goes away from the cutoff frequencies, the imaginary part of the saddle point \(\tau_n\) increases rapidly (see figure 1(g)). That means small quantum tunneling rate and hence weak sidebands. When the laser detuning is positive and \(\Delta \geq 3.17U_p\), the initial velocity of the e–h pair generated by real excitation is so large that the THz field cannot bring the electron and the hole to the same position again for recombination. The saddle-point solutions are complex. The corresponding saddle-point solutions of \(\tau\) are depicted in figures 1(h) and (i). When the emission frequency goes away from the excitation frequency or the band edge, the imaginary parts of the saddle points of \(\tau\) (figure 1(i)) increases rapidly, so
Figure 1. The dimensionless sideband frequency shift $N\omega_0/U_p$ in the plateau region as a function of the return time $\tau$ for various NIR laser detuning $\Delta = \Omega - E_g$. (a) $\Delta = 0.00$; (b) $\Delta = 0.20U_p$; (c) $\Delta = 0.91U_p$; (d) $\Delta = 2.20U_p$; (e) $\Delta = 3.09U_p$; (f) and (g) $\Delta = -0.09U_p$; (h) and (i) $\Delta = 4.41U_p$. The dots and crosses in (a)–(e) are for initial relative velocity of the e–h pair, anti-parallel and parallel to the THz field $E(t)$, respectively. In (f)–(i), the saddle-point solutions of $\tau$ are complex numbers and the results are plotted as functions of the real part of $\tau$. The frequency and strength of the THz field are 5 meV and 30 kV cm$^{-1}$, respectively. The reduced mass of the e–h pair is set as 0.076 $m_e$. These parameters give $U_p = 90$ meV.

The sideband strength $P_N$ decreases rapidly and presents sharp peaks at the band edge and the excitation frequency (see figure 2(b)).

We now show the quantum trajectory approach to calculating the HSG spectrum. By solving the saddle-point equations above, we obtain the saddle points $(t_n, \tau_n)$. The action is
Figure 2. HSG intensity as a function of excitation and emission frequencies. (a) The contour plot of the sideband strength $|\chi_N|^2$ as a function of detuning $\Delta = \Omega - E_g$ and sideband frequency $\omega$. Here the sideband strength is exactly calculated by numerical solution of equation (2). The width of the sideband plateau depends on $\Delta$. (b) The sideband strength $|\chi_N|^2$ calculated by the saddle-point method (triangles) and exactly (dots) with detuning above the $3.17U_p$ threshold ($\Delta = 397$ meV $\approx 4.41U_p$). The parameters are the same as in figure 1.

then

$$S_{cl}(t, \tau) = N \omega_0 t + (\Omega - E_g - 2U_p)\tau + 2U_p\tau \gamma^2 + 2U_p\tau \alpha \gamma \cos(2\omega_0 t - \omega_0 \tau),$$

(9)

where $\gamma = \sin(\omega_0 \tau/2)/(\omega_0 \tau/2)$. After integrating the polarization strength $P_N$ with Gaussian integral, the susceptibility is determined by [9]

$$\chi_N(\Omega + N\omega_0) = \frac{\omega_0 P_N(\Omega + N\omega_0)}{\pi E_{NIR}}$$

$$\approx \frac{\omega_0}{\pi} \sum_n \frac{i d^* d}{(\sqrt{4\pi \tau_n 1 + 0^+})^3} \exp[i S_{cl}(t_n, \tau_n)] \sqrt{\frac{4\pi^2 i^2}{\det S_{cl}''}}.$$  

(10)

Here $S_{cl}''$ represents the second-order derivative Jacobian determinant of the corresponding action [5]. The results calculated by exact solution and the saddle-point method are shown in figure 2. The exact solution of $\chi_N$ is obtained by numerically integrating equation (2). The characteristic of the HSG spectrum agrees well with the results in figure 1: the plateau shrinks when the detuning is increased. The saddle points $\tau_n$ and $t_n$ are complex outside the plateau region, which indicates that the quantum tunneling occurs in the emission. The lower cutoff frequency of the HSG plateau is the band edge of the semiconductor since the final kinetic energy of the e–h pair has to be positive. As discussed above, when the laser detuning is above the $3.17U_p$ threshold, the plateau feature vanishes and instead the sideband intensity presents two rapidly dropping peaks at the laser frequency and the band edge.
3. Summary

In summary, we have studied how high-order THz sideband generation in semiconductors driven by intense THz field depends on the detuning of the NIR laser from the semiconductor band edge. As compared with HHG in atoms, the HSG can be studied for positive laser detuning (laser above the semiconductor band edge), corresponding to unphysical ‘negative’ binding energy for HHG in atoms. Exact numerical simulation shows that for positive detuning, the HSG plateau shrinks with increasing laser detuning and eventually vanishes when the laser frequency is higher than the $3.17U_p$ threshold above the semiconductor band edge. Such features are well understood using the quantum trajectory approach.

Acknowledgments

This work was supported in part by Hong Kong RGC/GRF Project 401512 and the Hong Kong Scholars Program (grant no. XJ2011027). XTX thanks the National Natural Science Foundation of China (grant no. 61008016) and BFZ thanks the National Natural Science Foundation of China (grant no. 11074143).

Figure 3. The lower (stars) and upper (dots) cutoff frequencies of the sideband plateau as functions of the laser detuning $\Delta/U_p$. 

Figure 3 summarizes the cutoffs of the HSG plateau as functions of the laser detuning $\Delta$. By searching the real roots of the saddle points $\tau_n$ and $\tau_n$ for equations (8a) and (8b), we obtain the lower and upper cutoffs of the HSG spectrum. As shown in figure 3, the width of the HSG plateau decreases with the increasing of the laser detuning $\Delta$. But the variation is rather slow in the range $\Delta/U_p \in (0, 2.2)$.
Appendix. Motion of an e–h pair under a THz field

The e–h pair in our case obeys the inhomogeneous Schrödinger equation in the effective mass approximation

$$i\hbar \frac{\partial}{\partial t} \psi(r_e, r_h, t) = \left(-\frac{\hbar^2}{2m_e} \Delta_{r_e} - \frac{\hbar^2}{2m_h} \Delta_{r_h} + E_g - \frac{e^2}{4\pi\varepsilon_0 \varepsilon |r_e - r_h|} \right) \psi(r_e, r_h, t)$$

$$- \left[ eF \cos(\omega_0 t + k_0x_e)z_e - eF \cos(\omega_0 t + k_0x_h)z_h \right] \psi(r_e, r_h, t)$$

$$+ d \cdot E_L \cos(\Omega t + k_Lx_e) \delta(r_e - r_h),$$

where $m_e$ and $m_h$ denote the effective masses of the electron and the hole, respectively, $\delta(r_e - r_h)$ is a 3D delta function, which means that the electron and the hole are generated at the same position ($r_e = r_h$), $d$ is the electric dipole moment, $\varepsilon_0$ is the vacuum dielectric constant, and $\varepsilon$ is the relative background dielectric constant of the semiconductor. $F$ ($E_L$) and $\omega_0$ ($\Omega$) are the amplitude and frequency of the THz (NIR laser) field, respectively. In this paper, we consider optically thin samples (thickness $\ll$ wavelengths) and therefore the propagation effects (such as phase matching) are not important. Also, the optical wavelength ($\sim 1000$ nm) is much greater than the exciton radius ($\sim 10$ nm), so the dipole approximation is well justified [12]. With the assumption of optically thin samples and the dipole approximation, we can neglect the spatial dependence of the NIR and THz fields ($k_L$ and $k_0$ are set to be zero in equation (A.1)).

Using the center-of-mass and relative coordinates ($\mathbf{R} = \frac{m_e r_e + m_h r_h}{m_e + m_h}$ and $\mathbf{r} = r_e - r_h$) of the e–h pair with the corresponding total mass ($M = m_e + m_h$) and reduced mass ($\mu = \frac{m_e m_h}{m_e + m_h}$), the equation can be written as

$$i\hbar \frac{\partial}{\partial t} \Psi(\mathbf{R}, \mathbf{r}, t) = \left[-\frac{\hbar^2}{2M} \Delta_{\mathbf{R}} - \frac{\hbar^2}{2\mu} \Delta_{\mathbf{r}} + E_g \right] \Psi(\mathbf{R}, \mathbf{r}, t)$$

$$- \left[ eF \cos(\omega_0 t) z + \frac{e^2}{4\pi\varepsilon_0 \varepsilon |\mathbf{r}|} \right] \Psi(\mathbf{R}, \mathbf{r}, t) + d \cdot \mathbf{E}(t) \delta(\mathbf{r}),$$

(A.2)

where the rotating-wave approximation has been applied and $E(t) = E_L \exp(-i\Omega t)/2$. The center-of-mass and relative motions can be separated so that the wavefunction can be written as $\Psi(\mathbf{R}, \mathbf{r}, t) = \Phi(\mathbf{R}, t) \Psi(\mathbf{r}, t)$, which leads to the equation of relative motion for the e–h pair:

$$i\hbar \frac{\partial}{\partial t} \Psi(\mathbf{r}, t) = \left[-\frac{\hbar^2}{2\mu} \Delta_{\mathbf{r}} + E_g - eF \cos(\omega_0 t) z - \frac{e^2}{4\pi\varepsilon_0 \varepsilon} \right] \Psi(\mathbf{r}, t) + d \cdot \mathbf{E}(t) \delta(\mathbf{r}).$$

(A.3)

The Bohr radius and Rydberg energy of the e–h pair are $a_B = \frac{4\pi\varepsilon_0 \hbar^2}{e^2 \mu}$ and $E_{\text{Ryd}} = \frac{\hbar^2}{2a_B^2}$, respectively. For GaAs, we take $a_B = 10$ nm and $E_{\text{Ryd}} = 5$ meV. Using the Rydberg unit system (in which $2\mu$, $e^2/\hbar$, $h$, $a_B$ and $E_{\text{Ryd}}$ are all taken as unity), we obtain

$$i \frac{\partial}{\partial t} \Psi(\mathbf{r}, t) = \left[-\Delta_{\mathbf{r}} + E_g - F \cos(\omega_0 t) z - \frac{2}{r} \right] \Psi(\mathbf{r}, t) + d \cdot \mathbf{E}(t) \delta(\mathbf{r}).$$

(A.4)

The corresponding equation of motion in the velocity gauge then reads

$$i \frac{\partial}{\partial t} \Psi(\mathbf{r}, t) = \left[\left| \mathbf{p} - \mathbf{A}(t) \right|^2 + E_g - \frac{2}{r} \right] \Psi(\mathbf{r}, t) + d \cdot \mathbf{E}(t) \delta(\mathbf{r}),$$

(A.5)

where $\mathbf{A}(t) = -\frac{F}{\omega_0} \sin(\omega_0 t) \hat{z}$ is the vector potential of the intense THz field.
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