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Femtosecond chirped pulse control of photoluminescence and generating electron–hole pairs in broadband semiconductors

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Abstract

We have studied an intense chirped pulse excitation of broadband semiconductors with a direct interband transition. We concentrate on photoluminescence and generating electron–hole pairs. The important role of the pulse frequency modulation for obtaining high excited carriers' densities has been demonstrated. We show that a mechanism of generating electron–hole pairs strongly depends on the chirp rate in the frequency domain $\Phi''(\nu)$. As $\Phi''(\nu)$ increases, a behavior related to the Rabi oscillations of the carriers' densities gives way to adiabatic rapid passage and thereafter to an incoherent excitation of carriers. Correspondingly, the carriers' densities strongly increase when the behavior related to the Rabi oscillations gives way to adiabatic rapid passage. In the incoherent regime the excited carrier densities vary slower, and the carrier–carrier scattering has severe effects on their behavior in this region.

Keywords: Chirped pulse; Ultrafast processes; Broadband semiconductors; Coherent excitation of carriers

1. Introduction

Selective population transfer with phase modulated (chirped) pulses has applications in a number of areas, such as preparation of initial states for spectroscopy, optical quantum control of atoms and molecules [1] and Bose–Einstein condensates. In this work we concentrate on photoluminescence and generating electron–hole pairs in broadband semiconductors with a direct interband transition. Such an investigation is initiated also by an experimental study by Kunde et al. [2] of the influence of frequency chirp on broadband semiconductor continuum nonlinearities. Pulse chirping has the potential to improve and optimize all-optical ultrafast switching.

Optical excitation of broadband semiconductors has some similarities to optical transitions in molecules. First, the Franck–Condon principle in molecules is similar to the momentum conservation in direct semiconductors. Second, in the absence of relaxation the Bloch equations for both the molecular system [3] and the broadband semiconductor [4,5] describe an ensemble of independent two-level systems with different transition frequencies corresponding to a pure inhomogeneously broadened optical transition.

The possibility of controlling molecular dynamics using properly tailored pulses has been the subject of intensive studies in the last few years. Chirped pulses can selectively excite coherent wave

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packet motion either on the ground electronic potential energy surface of a molecule or on the excited electronic potential energy surface due to the intrapulse pump–dump process [6,7]. In addition, such pulses are very efficient for achieving population transfer between molecular electronic states.

Two well-known procedures based on a coherent excitation can, in principle, produce complete population inversion in an ensemble of two-level atoms. The first one is the π-pulse excitation [1]. This method makes use of the Rabi population oscillation. The main disadvantage of the π-pulse excitation is the requirement for resonant laser light and the need of precise control of the pulse area [1]. The second procedure, known as adiabatic rapid passage (ARP) [1], is based on sweeping the pulse frequency through a resonance. A scheme based on ARP is robust since it is insensitive to pulse area and to the precise location of the resonance.

Electronic population transfer in molecules is in general much more complicated than in atoms due to the nuclear motion influence. The intrapulse pump–dump process and the ARP by intense ultrashort chirped pulses in molecules coupled with a dissipative environment have been studied in Refs. [8,3], respectively. By this means one would expect an effective control of broadband semiconductors using intense chirped pulses by analogy with molecules.

We have calculated the carrier densities (of electron and holes) excited by intense ultrashort chirped pulse after the completion of the pulse action. The present investigation is carried out for semiconductors with a direct interband optical transition by solving semiconductor Bloch equations [4,5]. These equations describe both the coherent excitation and incoherent processes [5]. In our study attention is focused on different mechanisms of the coherent optical excitation of carriers: the π-pulse excitation and the ARP.

Following Ref.[4] we consider high excited carriers' densities, therefore to a first approximation, one can neglect coherent many body effects like the band gap renormalization due to the Coulomb exchange interaction and the Coulomb electron–hole correlations. However, we do take into consideration incoherent many-body effects like the carrier distribution function relaxation and dephasing the optical polarization due to carrier–carrier scattering.

2. Basic equations

The semiconductor is affected by phase modulated pulse of carrier frequency ω

$$E(t) = \frac{1}{2}\varepsilon(t)\exp(-\mathrm{i}\omega t + \mathrm{i}\varphi(t)) + \mathrm{c.c}, \tag{1}$$

where $\varepsilon(t)$ and $\varphi(t)$ are the real functions of time, and $\varphi(t)$ describes the change of the pulse phase in a time t. The instantaneous pulse frequency is $\omega(t) = \omega - \mathrm{d}\varphi(t)/\mathrm{d}t$.

We describe an ultrafast laser–semiconductor interaction by the semiconductor Bloch equations [4]. Below these equations are written for the distribution functions of the electrons F_p^{e}, holes F_p^{h} and the negative frequency component of the polarization $\mathsf{P}_p = P_p \exp(\mathrm{i}\omega t - \mathrm{i}\varphi(t))$ in the interaction representation:

$$\frac{\mathrm{d}F_p^{\mathrm{c}}}{\mathrm{d}t} = -\mathrm{i}\frac{1}{2}U_p^*\mathsf{P}_p + \mathrm{i}\frac{1}{2}U_p\mathsf{P}_p^* - \Gamma_p^{\mathrm{c}}(F_p^{\mathrm{c}} - f_p^{\mathrm{c}}(\mu_{\mathrm{c}}(t), T(t))), \tag{2}$$

$$\frac{\mathrm{d}\mathrm{P}_p}{\mathrm{d}t} = (\mathrm{i}\Delta_p - \gamma_p)\mathsf{P}_p + \mathrm{i}\frac{1}{2}U_p(1 - F_p^{\mathrm{e}} - F_p^{\mathrm{h}}). \tag{3}$$

Here the quantity $U_{\mathrm{p}} = \Omega_{\mathrm{R}} = \mathrm{d}_{cv}\varepsilon(t)/\hbar$ is equal to the Rabi frequency of the semiconductor in our approximation, $\Delta_p = \Delta_0 - \mathrm{d}\varphi(t)/\mathrm{d}t - \varepsilon_p^0/\hbar, \Delta_0 = \omega - E_{\mathrm{g}}/\hbar$ is the detuning of the carrier frequency ω with respect to the semiconductor band-gap E_{g}, ε_p^0 is the kinetic energy of the electron–hole pair; c= e, h

As to the scattering terms for the carrier distribution functions (see the last term on the right-hand side of Eq. (2)), we use the relaxation rate approximation [4]. According to this approximation, the population-scattering processes tend to distribute the electrons and holes in quasiequilibrium Fermi distributions $f_{\mathrm{p}}^{\mathrm{c}}(\mu_{\mathrm{c}}(t), T(t))$ with chemical potential $\mu_{\mathrm{c}}(t)$ and carrier temperature $T(t)$. Two last quantities are fixed by the conservation of the total particle number and the total kinetic energy in the carrier–carrier scattering

process

$$n_c = 2\sum_p F_p^c, \langle \varepsilon_{kin} \rangle = 2\sum_{p,c} \varepsilon_p^c F_p^c. \quad (4)$$

Quantities Γ_p^e and Γ_p^h are the relaxation rates of the electrons and holes, respectively. The dephasing of polarization is described with a rate term $\gamma_p = \gamma_p^{sc} + \gamma_p^0$, where the first term $\gamma_p^{sc} = (\Gamma_p^e + \Gamma_p^h)/2$ takes into account the contribution from the relaxation rates of the electrons and holes, and the second term γ_p^0 takes into account all other mechanisms [4]. Usually the second term is much smaller than first one: $\gamma_p^0 \ll \gamma_p^{sc}$.

Following Ref. [9], we take into account the dependence of the matrix element of the dipole moment on the carrier energy due to unparabolicity of the band structure

$$d_{cv}(\varepsilon_p^0) = \frac{d_{cv}}{1 + \exp((\varepsilon_p^0 - \varepsilon_c)/\Delta_c)} \quad (5)$$

where $\Delta_c = 1.05$ mev and $\varepsilon_c = 270$ mev for GaAs. Such a dependence leads to the limitation on the energy of the carriers' excitation.

3. Results and discussions

The calculation results have been obtained by numerical solution of Eqs. (2)–(5) for a Gaussian pulse of the shape

$$E(t) = \frac{E_0}{2}\left(\exp\left[- i\omega t.. - \frac{1}{2}(\delta^2 - i\mu)(t - t_0)^2\right] + c.c\right). \quad (6)$$

If chirped pulses are obtained by changing the separation of pulse compression gratings, the parameters δ and μ are determined by the formulae

$$\delta^2 = \left\{\tau_0^2 + \left[\frac{\Phi''^2(\omega)}{\tau_0^2}\right]\right\}^{-1}, \quad \mu = -\Phi''(\omega)[\tau_0^4 + \Phi''^2(\omega)]^{-1} \quad (7)$$

where $\tau_0 = t_{p0}/2\sqrt{\ln 2}$, t_{p0} is the duration of the transform-limited pulse, $\Phi''(\omega) = \Phi''(\nu)/4\pi^2$, and $\Phi''(\nu)$ is the chirp rate in the frequency domain.

All the calculations were performed for an optically thin bulk sample of GaAs at room temperature. The values of parameters were taken from Refs. [4,5]. We put $\Gamma_p^e = \Gamma_p^h = 1/60\ fs^{-1}$. The exciting pulse parameters were taken as the following: $t_{p0} = 13$ fs, the peak intensity was 10^8–10^9 W/cm^2 in the order of magnitude. Initial values of the carrier densities were equal to zero: $n_e = n_h = 0$.

Fig. 1 shows the influence of the dimensionless pulse area $S_0 = \int_{-\infty}^{+\infty} d_{cv}\mathsf{E}(t)/\hbar\, dt$ on the excited carriers' densities n_e and n_h, and the luminescence signal that is proportional to the product of the electron and hole densities $n_e n_h$ after the completion of the pulse action for zero chirp. Our results display a behavior related to the Rabi oscillations. We observe maxima of the excited carriers densities for the values of the dimensionless pulse area S_0 which are close to π, 3π,… Correspondingly one can see minima of the excited carriers densities for $S_0 = 2\pi, 4\pi$,… The values of the carriers' densities in extreme points increase with the pulse area. Such a behavior can be explained using an analytical solution for the sech-like pulse $E = E_0\,\mathrm{sech}((t - t_0)/\tau))$ without phase modulation and in the absence of relaxation [10], as applied to our model. Taking into account the dipole moment dependence on the carriers' energy due to limitation of the excitation range for $\varepsilon_c \gg \Delta_c$ (see Eq. (5)), and the density of states in a semiconductor, we get the following equation for the sum of the excited carrier densities after the pulse completion:

$$n_e + n_h = \frac{2\sqrt{2}m_{eh}^{3/2}}{\pi^2\ \hbar^3} \sin^2\left(\frac{S_0}{2}\right)\int_0^{\varepsilon_c} d\varepsilon\sqrt{\varepsilon}\frac{1}{\cosh^2\left(\pi\tau/2(\varepsilon/\hbar - \Delta_0)\right)}. \quad (8)$$

Eq. (8) describes the dependence on the pulse area S_0 which is similar to that of the atomic system. If $\Delta_c \sim \varepsilon_c$, one can expect extreme points for different values of pulse areas. Fig. 2 shows the influence of the pulse energy flux on the excited carriers' densities n_e and n_h , and the luminescence signal ($\sim n_e\ n_h$) after the completion of the pulse action for chirped pulses. One can see the oscillations still persist at moderate chirp rates in the frequency domain when $\Phi''(\nu) < 10{,}000\ fs^2$. The larger is the chirp rate, the smaller is the oscillation amplitude.

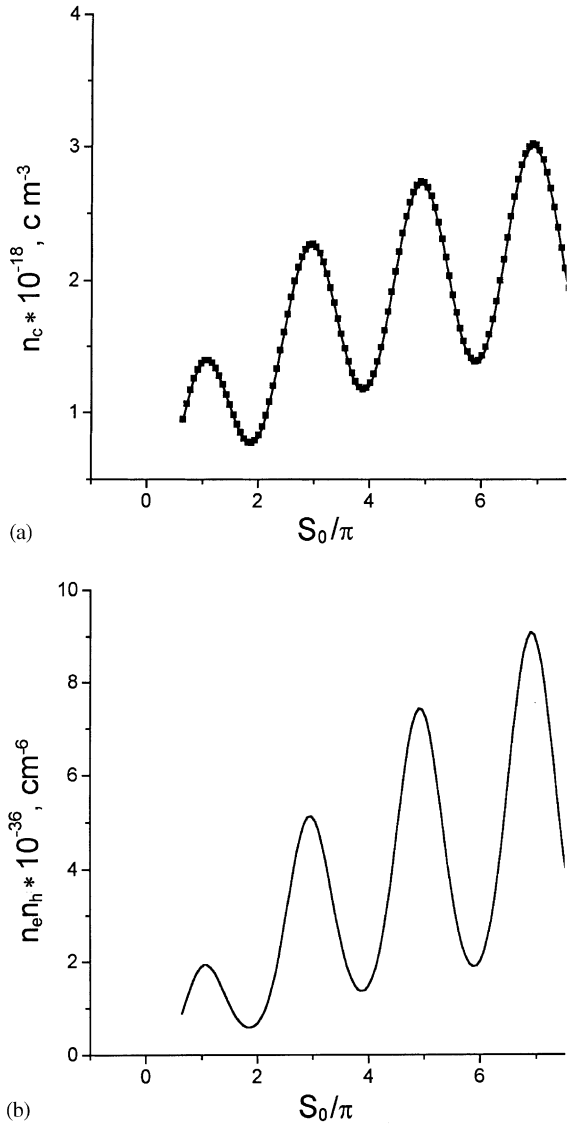

Fig. 1. (a) The excited carrier densities n_e (solid line) and n_h (dotted line), and (b) the luminescence signal $\sim n_e n_h$, after the completion of the pulse action as the functions of pulse area for zero chirp ($\mu = 0$) and zero frequency detuning ($\Delta_0 = 0$).

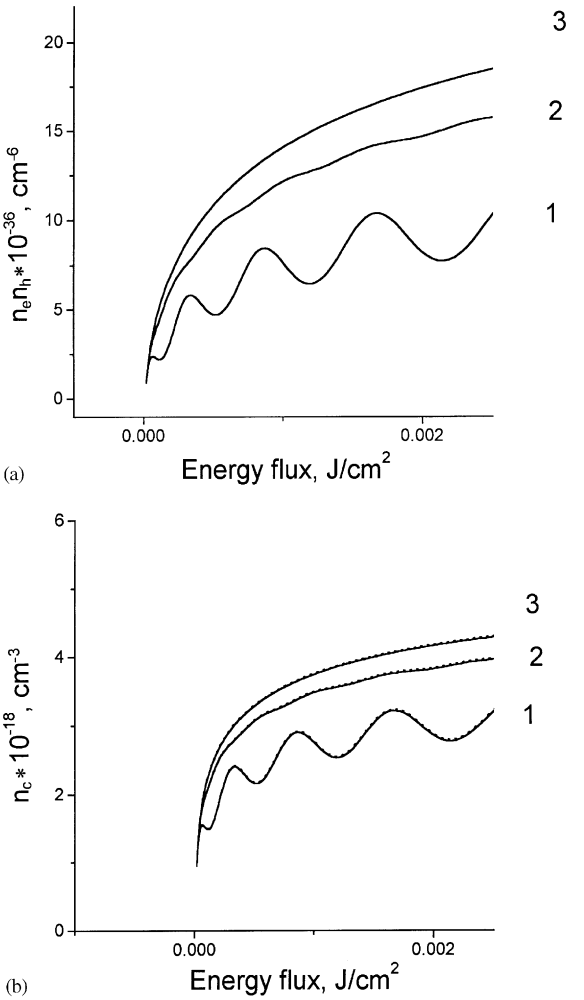

Fig. 2. (a) The excited carrier densities n_e (solid line) and n_h (dotted line), and (b) the luminescence signal $\sim n_e n_h$, after the completion of the pulse action as the functions of pulse energy flux for $\Delta_0 = 0$. $\Phi''(v) = 2000\,\mathrm{fs}^2$(1), $5000\,\mathrm{fs}^2$(2) and $10{,}000\,\mathrm{fs}^2$ (3).

The oscillations disappear for $\Phi''(v) \geqslant 10{,}000\,\mathrm{fs}^2$, and one obtains a smooth increase in n_e and n_h with the pulse energy flux.

Figs. 3 and 4 show the calculation results of the excited carriers' densities n_e and n_h , and the luminescence signal as functions of $\Phi''(v)$ for different pulse energy fluxes corresponding to extreme pulse areas S_0 (for zero chirp).

One can see that with a rise in the absolute value of $\Phi''(v)$ in the limit of $\Phi''(v) < 10{,}000\,\mathrm{fs}^2$, the carriers' densities increase. Such an increase is strong for pulse areas S_0 corresponding to minima in Fig. 1, and essentially weaker for the values of S_0 corresponding to maxima. For larger values of $\Phi''(v) > 10{,}000\,\mathrm{fs}^2$, one can observe smooth variations of the carriers' densities.

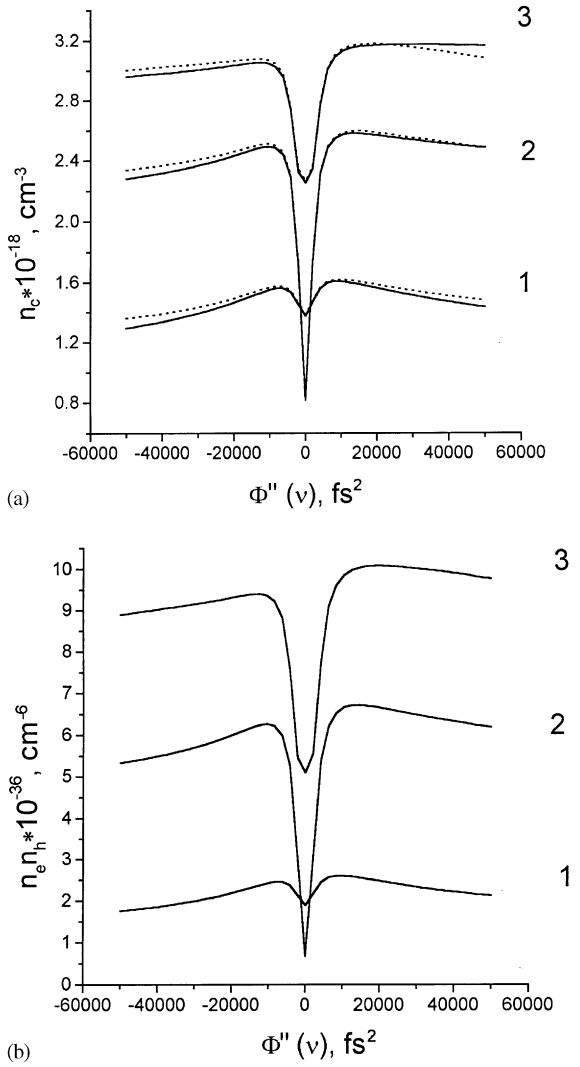

Fig. 3. (a) The excited carrier densities n_e (solid line) and n_h (dotted line), and (b) the luminescence signal $\sim n_e n_h$, after the completion of the pulse action as functions of $\Phi''(\nu)$ for $\Delta_0 = 0$. The corresponding pulse areas at zero chirp S_0 are equal to π (1), 2π (2) and 3π (3).

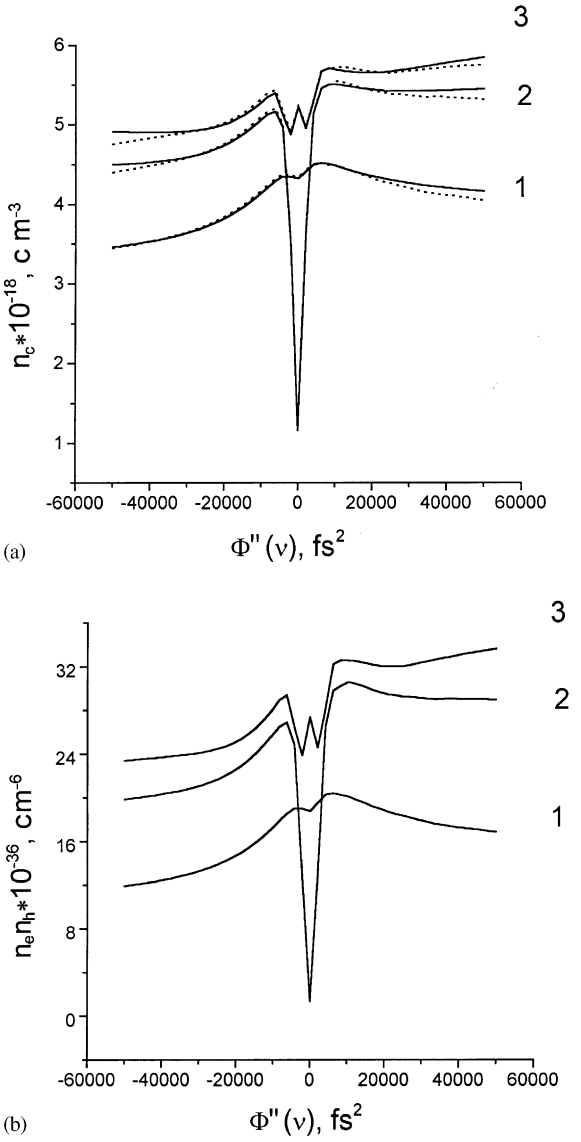

Fig. 4. The same as Fig. 3 with the only difference that Δ_0 =84.3 mev.

Figs. 3 and 4 display the larger is the frequency detuning Δ_0, the larger is the asymmetry in the carrier densities' dependence on the chirp sign. The densities are higher for positive chirp.

To understand the behavior described before, we will discuss first our model in the absence of relaxation. Then it is an ensemble of noninteracting two-level systems with different transition frequencies. The values of chirp rates in the frequency domain $\Phi''(\nu) \geqslant 10{,}000\,fs^2$ correspond to the limit of strongly chirped pulses when the pulse duration is much larger than that of the transform limited one. Then (see Ref. [3]) $|\Phi''(\omega)| \gg \tau_0^2$. For such a condition the ARP

criterion for a two-level system is the following:

$$\left|\frac{d\omega(t)}{dt}\right| \ll \Omega_R^2(t). \quad (9)$$

For linear chirped pulses determined by Eqs. (6) and (7), we obtain by Eq. (9)

$$S_0 \gg \sqrt{2\pi}. \quad (10)$$

One can see that this criterion holds for the values of S_0 used in our calculations. By this means the behavior shown in Figs. 3 and 4 in the region of $|\Phi''(\nu)| \sim 10{,}000\,\mathrm{fs}^2$ can be explained by the ARP. For larger values of $\Phi''(\nu)$, the pulse duration increases and becomes larger than the values of relaxation times. Under these conditions a coherent regime of ARP gives way to an incoherent behavior.

4. Conclusion

In this work we have studied the phase modulation effects on generating electron–hole pairs (n_e and n_h) and photoluminescence in a broadband semiconductor excited with intense ultrashort chirped pulse. Our calculation results display a behavior related to the Rabi oscillations of n_e and n_h in the absence of the pulse chirp. When the pulse chirp is not zero, the oscillations still persist at moderate chirp rates in the frequency domain $\Phi''(\nu)$. The oscillations disappear for $\Phi''(\nu) \geqslant 10{,}000\,\mathrm{fs}^2$. In addition, we observed a strong dependence of n_e and n_h on the value of the chirp rate in the frequency domain $\Phi''(\nu)$, its sign and the detuning Δ_0 of the carrier pulse frequency ω with respect to the semiconductor band-gap E_g. The larger is the frequency detuning, the larger is the asymmetry in the carrier densities' dependence on the chirp sign.

According to our consideration, as $\Phi''(\nu)$ increases, a behavior related to the Rabi oscillations of n_e and n_h gives way to ARP and thereafter to an incoherent excitation of carriers.

In the near future we intend to include in our consideration the effects of the Coulomb electron–hole correlations.

Acknowledgements

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