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Time-delayed behaviors of transient four-wave mixing signal intensity in inverted semiconductor with carrier-injection pumping

Zhenhua Hu *, Shen Gao, Bowen Xiang

Department of Physics and Technology, School of Science, Wuhan University of Technology, Wuhan 430063, China

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ABSTRACT

An analytical expression of transient four-wave mixing (TFWM) in inverted semiconductor with carrier-injection pumping was derived from both the density matrix equation and the complex stochastic stationary statistical method of incoherent light. Numerical analysis showed that the TFWM decayed decay is towards the limit of extreme homogeneous and inhomogeneous broadenings in atoms and the decaying time is inversely proportional to half the power of the net carrier densities for a low carrier-density injection and other high carrier-density injection, while it obeys an usual exponential decay with other decaying time that is inversely proportional to half the power of the net carrier density or it obeys an unusual exponential decay with the decaying time that is inversely proportional to a third power of the net carrier density for a moderate carrier-density injection. The results can be applied to studying ultrafast carrier dephasing in the inverted semiconductors such as semiconductor laser amplifier and semiconductor optical amplifier.

1. Introduction

Transient four-wave mixing (TFWM) [1], which acts as a kind of nonlinear laser spectroscopy with ultrahigh time-resolution, plays a significant role in studying dephasing and population relaxation times of atoms as well as vibration dephasing and Kerr relaxation of molecules in the femtosecond–picosecond regime [2]. Also it as a Doppler-free nonlinear laser spectroscopy is applied to measurement of the energy-level splitting in atoms [3–7]. Nowadays it has become the important method for detecting the ultrafast dephasing time of semiconductor optoelectronic materials, such as bulk [8–10], quantum well [11,12] and quantum dot [13,14] structured semiconductors.

The TFWM signal intensity is usually expressed by the time integration of function of the field amplitude (or intensity) and the response function of matter and the time-resolution is usually determined by the correlation time associated with incoherent fluctuating light instead of temporal duration of pulsed laser. For the atoms with two-level, the delayed signal at resonance is exponentially decayed with constants $2\gamma_2$ and $4\gamma_2$ for an extreme homogeneous and other inhomogeneous broadenings, respectively, where γ_2 is the dephasing rate of atoms [1]. However, the situations are changed in semiconductors because there are additional intraband ultrafast carrier relaxations, such as carrier–carrier scattering, carrier–LO–phonon scattering, Coulomb screening effect and so on. In the earlier work, Becker showed experimentally the decay of the delayed signal is closely related to both quantum kinetics and background carrier density (BCD), N, and the dephasing rate satisfies the relation $\propto N^{1/3}$ [8]. Accordingly, an unusual delayed decay of the TFWM signal was found by Vu et al., which obeys the law $\propto \exp(-aN\tau^3)$ when the excited resonantly pulse is shorter than the built-up time of the Coulomb screening [9]. The delayed decays occur in the inverted semiconductors with light pumping [15–17] as well as with carrier-injection pumping [18,19]. The results all showed that the delayed behaviors are considerably different from those of atoms.

In the matter of the inverted semiconductors with carrier-injection pumping such as semiconductor laser amplifier (SLA) and semiconductor optical amplifier (SOA), the dephasing processes are quite complicated because it refers to the carrier, phonon and Coulomb interactions. To our knowledge, no such delayed decay law of the TFWM signal has been used for studying the ultrafast dephasing time in the active gain semiconductor up to now. In this paper, we will develop a theory to describe the delayed behaviors of the TFWM signal for the active gain semiconductors. In our calculations, our model is based on the two direct-band structures with carrier-injection pumping and the TFWM configuration is considered to be a phase-conjugate geometry, as well as the contribution of the gain spectrum with a parabolic profile is included. Our calculations showed that the delayed decays of three cases can occur in the active gain semiconductors. That is, the TFWM signal is towards the delayed decays $I(\tau)\propto\exp(-2\gamma_{\mathbf{k}2}\tau)$ and

* Corresponding author.
E-mail address: z.h.hu.phys.dept@whut.edu.cn (Z.H. Hu).

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$\propto \exp(-4\gamma_{\mathbf{k}2}\tau)$ for a low carrier-density injection and other high carrier-density injection, respectively, while it obeys another unusual law for a moderate carrier-density injection.

The paper is organized as follows. Section 2 is the model and theory of the TFWM signal with a phase-conjugate geometry. Section 3 is a general analysis for the TFWM signal. In Section 3 we present the conclusions.

2. Model and theory

The band-structured model of the active gain semiconductor with conduction-band state, $|\mathbf{k}, c\rangle$, and valence-band state, $|\mathbf{k}, v\rangle$, is shown in Fig. 1(a). The electronic and hole's quasi-Fermi energy levels F_c and F_v are shifted into the conduction- and valence-bands, respectively, as a result of carrier-injection pumping. The population is inverted and an optical gain is yielded when the quasi-Fermi energy-level difference $F_c - F_v$ of the semiconductors is larger than the bandgap E_g. The gain spectrum is [20]

$$G(\Delta\omega) = \Gamma\left[a_1(N - N_0) - a_2\hbar^2(\omega' - \omega'_N)^2\right]. \tag{1}$$

where N and N_0 are the BCD and the transparency carrier density, respectively, Γ is the confinement factor of active region, a_1 is the gain factor, a_2 is the constant related to material, and $\omega'_N = \omega'_0 + a_3(N - N_0)$ is the peak angular frequency of gain spectrum (ω'_0 is the peak angular frequency of gain spectrum at transparency point and a_3 is the constant of frequency shift). The full width at half maximum (FWHM), $\delta\Omega$, satisfies

$$\delta\Omega = \sqrt{2}\beta, \tag{2}$$

where

$$\beta = \sqrt{\frac{a_1(N - N_0)}{a_2\hbar^2}} \tag{3}$$

is the positive root of the zero gain. Apparently, there only is the homogeneous broadening determined by the BCD instead of inhomogeneous one caused by the Doppler's effect unlike those in atomic vapor. In general, the spectrum is responsible for gain and phase shift due to band filling as well as responsible for bandgap renormalization due to Coulomb screening effect. Currently the parabolic gain have been widely applied to studying linear and nonlinear optical characteristics of the active semiconductors due to desiring of the optical telecommunications, such as all-optical data process and wavelength conversion based on semiconductor laser amplifiers (SLA) and semiconductor optical amplifiers (SOA) [21–27]. We will reveal here the delayed behaviors of the TFWM signal associated with carrier–carrier scattering, carrier–LO–phonon scattering and plasma oscillation to apply the TFWM to studying the nonlinear polarization response of the active gain semiconductors.

The TFWM is considered to be the phase-conjugate geometry shown in Fig. 1(b). The resonant pump beams 1 and 2 are the temporally incoherent lights that come from a single broadband laser source, whose central frequency, ω_0, is centered at ω'_N, and a small angle exists between them; another resonant probe beam 3 is the coherent light that comes from quasi-monochromatic laser source with angular frequency ω_0 is almost propagating opposite to beam 1. In this case, the total light field interacting with the active gain semiconductors can be expressed as

$$\begin{aligned} E(\mathbf{r}, t) = {} & \epsilon_1 R(t)\exp[i(\mathbf{K}_1\cdot\mathbf{r} - \omega t)] \\ & + \epsilon_2 R(t + \tau)\exp\{i[\mathbf{K}_2\cdot\mathbf{r} - \omega(t + \tau)]\} \\ & + \epsilon_3 \exp\{i[\mathbf{K}_3\cdot\mathbf{r} - \omega_0 t]\} + \text{c. c.}, \end{aligned} \tag{4}$$

where $\mathbf{K}_i$ (i=1, 2, 3) is the wave vector of ith beam, ε_i is the

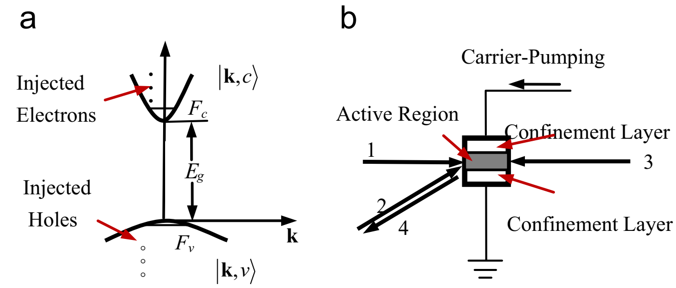

Fig. 1. (a) Energy band diagram of the active semiconductors with carrier-injection pumping and (b) schematic diagram of phase-conjugate geometry.

constant field amplitudes, τ is the time delays of beams 2 and 3, and $R(t)$ is a dimensionless factor that contains both phase and amplitude fluctuations [1,4–7]. For the parabolic bands shown in Fig. 1(a), the $\mathbf{k}$-dependent density matrix can be written as

$$\begin{aligned} \rho_{\mathbf{k}}(t) = {} & \rho_{\mathbf{k},c}(t)|\mathbf{k}, c\rangle\langle\mathbf{k}, c| + \rho_{\mathbf{k},v}(t)|\mathbf{k}, v\rangle\langle\mathbf{k}, v| \\ & + \rho_{\mathbf{k},cv}(t)|\mathbf{k}, c\rangle\langle\mathbf{k}, v| + \rho_{\mathbf{k},vc}(t)|\mathbf{k}, v\rangle\langle\mathbf{k}, c|, \end{aligned} \tag{5}$$

where $\rho_{\mathbf{k},c}(t)$ and $\rho_{\mathbf{k},v}(t)$ denote the electron and hole occupation probabilities in the conduction and valence bands, respectively, for an electron and hole state $\mathbf{k}$ in the active gain semiconductors, and $\rho_{\mathbf{k},cv}(t)$ $(=\rho_{\mathbf{k},cv}{}^*(t))$ is proportional to the interband polarization, and $\rho_{\mathbf{k}}(t)$ satisfies the equation of motion

$$\frac{\partial\rho_{\mathbf{k}}(t)}{\partial t} = \frac{i}{\hbar}\left[\rho_{\mathbf{k}}(t), H_0 + H_1(t)\right] + R_{\mathbf{k}}(t). \tag{6}$$

where H_0 is the unperturbed Hamiltonian, written as

$$H_0 = \left(\hbar\omega_{\mathbf{k},c} + E_g\right)|\mathbf{k}, c\rangle\langle\mathbf{k}, c| + \hbar\omega_{\mathbf{k},v}|\mathbf{k}, v\rangle\langle\mathbf{k}, v|, \tag{7}$$

where $\hbar\omega_{\mathbf{k},c}$ and $\hbar\omega_{\mathbf{k},v}$ are the quantum kinetic energies in the conduction-band and in the valence-band, respectively; $H_1(t)$ is the carrier-light interaction Hamiltonian, given by

$$H_1(t) = -\mu_{\mathbf{k}}E(\mathbf{r}, t), \tag{8}$$

where $\mu_{\mathbf{k}}$ is the dipole operator formed by the conduction-band and valence-band $\mathbf{k}$ states under the light fields. $R_{\mathbf{k}}(t)$ is the phenomenological term for describing the relaxation of the density matrix to the steady-state without light, read as [28,29]

$$R_{\mathbf{k}}(t) = -\gamma_{\mathbf{k}}^1[\rho_{\mathbf{k}}(t) - f_{\mathbf{k}}] - \gamma_{\mathbf{k}}^L[\rho_{\mathbf{k}}(t) - f_{\mathbf{k}}^L] - \gamma_{\mathbf{k}}^s[\rho_{\mathbf{k}}(t) - f_{\mathbf{k}}^{eq}]. \tag{9}$$

On the right hand side of (9), the first term is towards Fermi function $f_{\mathbf{k}}$ with rate $\gamma_{\mathbf{k}}{}^1$, the second term is towards Fermi function $f_{\mathbf{k}}{}^L$ of lattice temperature T_L with rate $\gamma_{\mathbf{k}}{}^L$, and the third term is towards temperature equilibrium function $f_{\mathbf{k}}{}^{eq}$ with rate $\gamma_{\mathbf{k}}{}^{eq}$. In other words, when the external light field interacts with the active semiconductors, the photoexcited carriers go through the dephasing relaxations caused by both carrier–carrier and carrier–LO scatterings as well as the energy relaxation due to quasi-Fermi energy level shift.

If $|H_{1cv}(t)|$ $(=|\langle\mathbf{k},c|H_1(t)|\mathbf{k},v\rangle|) \ll E_g$, where $H_{1cv}(t)$ is the Hamiltonian matrix element of the light field interacting resonantly with the active semiconductors, $H_{1cv}(t)$ can be regarded as a perturbation and $\rho_{\mathbf{k}}(t)$ can be expanded as a series form $\rho_{\mathbf{k}}(t) = \rho_{\mathbf{k}}{}^{(0)}(t) + \rho_{\mathbf{k}}{}^{(1)}(t) + \cdots + \rho_{\mathbf{k}}{}^{(n)}(t) + \cdots$ (n=0, 1, 2, ⋯,). Performing the perturb chain $\rho_{\mathbf{k}}{}^{(0)}(t) \to \rho_{\mathbf{k}}{}^{(1)}(t) \to \rho_{\mathbf{k}}{}^{(2)}(t) \to \rho_{\mathbf{k}}{}^{(3)}(t)$ and taking out only the phase-conjugate wave component with the conjugate-wave vector as $\mathbf{K}_4 = \mathbf{K}_1 - \mathbf{K}_2 + \mathbf{K}_3$ [2,30] the third-order interband density matrix elements are obtained as