

## EFFECT OF NONLINEAR WAVE MIXING ON ULTRAFAST MODULATION OF INTERBAND LIGHT IN SEMICONDUCTOR QUANTUM WELLS

*A. Neogi*

*New Energy Development Organization, FESTA Laboratories  
Tsukuba 300-26, Ibaraki, Japan*

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We investigate the effect of an induced sum-frequency signal on the transient modulation of interband-resonant probe light by a train of intersubband-resonant coupling light pulses in undoped quantum wells. The origin of the generated sum-frequency signal lies in the asymmetry of the three-level quantum well structure. The modulation characteristics are found to be significantly affected in the presence of a strong sum-frequency signal that builds up proportionately to the input probe field.

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Coherent optical effects in semiconductor devices are currently being extensively explored for their application in various information systems associated with generation, propagation, processing and detection of ultrashort signals. The investigation of nonlinear optical phenomena in multi-level quantum well structures involving the simultaneous utilization of both interband and intersubband transitions [1-10] have consequently attracted considerable attention. The study of transient properties of control of light by light in quantum wells provides an insight into its possible application in an optical switch in which the transmission of a highly absorptive medium can be controlled by an additional coupling light field. The transient analyses for a three-level quantum well system reveals that for resonantly interacting fields under ideal conditions the amount of probe absorption by the semiconducting medium act as the amplitude of a damped harmonic oscillator [5-8]. The ultimate modulation speed of interband (IB) light by intersubband (ISB) light in quantum wells is extremely fast as it depends on the intersubband relaxation rate [1]. Higher modulation speed can therefore be expected on application of ultrashort ISB coupling pulses in the femtosecond regime [6].

In this letter we investigate the modulation process in a three-level asymmetric quantum well system in the presence of an additional interband sum-frequency field [2] generated due to the nonlinear interaction of the interband probe and intersubband coupling light fields. Our recent analyses [6, 8] reveal that the modulation of a strong interband probe light by a train of coupling light pulses with pulse width and repetition rates comparable or longer than the intersubband life-times and interband relaxation rates is most efficient. The interband probe light is modulated on switching of a train of intersubband coupling light pulses at a correspondingly high speed, and depends critically on the intensities of the coupling and the probe light fields. We investigate the effect of the additional induced optical field on the transient absorption of the interband probe field by comparing the modulation process in both symmetric and asymmetric structures.

The density-matrix approach is employed for the study of this investigation. The steady-state modulation characteristics of the probe light by an intersubband

coupling light in an undoped quantum well can be clearly interpreted in terms of the excitonic effects, where the system becomes a completely discrete three-energy level system [1]. The interband- and intersubband-transitions are considered to correspond to two individual oscillators. The interband-related oscillator oscillates with its own frequency due to the resonant probe field in the absence of the coupling light. In the presence of a strong coupling field the intersubband-related oscillator becomes coupled to the interband-related oscillator. The strong coupling between the two oscillators leads to the deviation of the oscillation frequency from their own frequencies leading to a splitting of a single excitonic peak into two peaks. We consider this transient analysis at the resonance frequency where the reduction in the absorption of the probe field due to electromagnetically induced transparency by the coupling field is maximum. The three-level quantum well system is shown in Fig. 1. A cw interband probe light  $\{F_p \exp(-i\omega_p t) + \text{c.c. at } \omega_p\}$  is resonantly tuned to the first heavy-hole state-first conduction sub-band transition  $|a\rangle - |b\rangle$ . The ISB coupling field consists of a train of resonant pulses,  $\{[F_c \text{sech}(t/\tau_p) \exp(-i\omega_c t) + \text{c.c.}] \text{ at } \omega_c\}$  tuned to the transition  $|b\rangle$  and  $|c\rangle$  and is switched on at time  $t=0$  when the three-level system is at a steady state in presence of the cw probe field. The pulses are also assumed to be in phase, that is coherent with respect to one another. In an asymmetric quantum well structure there is an additional polarization at the sum-frequency field  $\omega_s = (\omega_p + \omega_c)$  arising from the finite transition dipole moment ( $\mu_{ca}$ ) between the first heavy-hole state  $|a\rangle$  and the second conduction subband state of the three-level system  $|c\rangle$ . This allows a direct interband transition between the states  $|c\rangle$  and  $|a\rangle$  which is otherwise forbidden under the interband selection rules in a symmetrical structure where the dipole moment  $\mu_{ca}$  is rendered zero. Thus the modulation process in symmetric wells involves two-wave interaction with frequencies  $\omega_p$  and  $\omega_c$  while in asymmetric well it involves the interaction of three distinct waves at  $\omega_p$ ,  $\omega_c$  and  $\omega_s$ . This induced sum-frequency signal  $\{F_s(t) \exp(-i\omega_s t) + \text{c.c.}\}$  is expected to modify the modulation characteristics of the interband light.

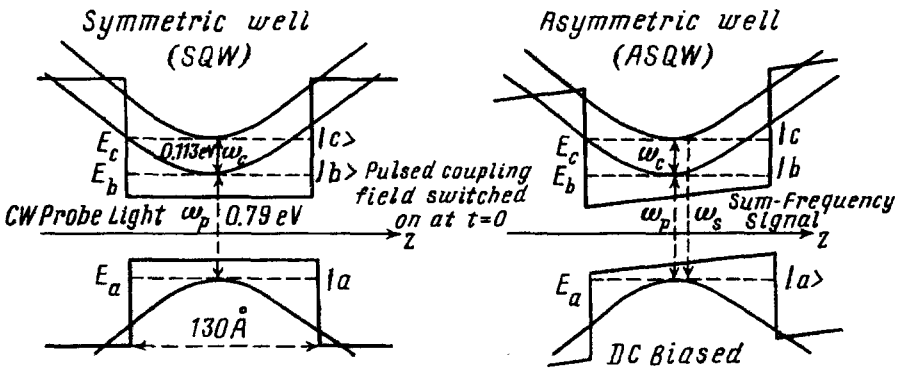


Fig.1. Schematic drawing for modulation of interband resonant light by intersubband resonant light in undoped quantum wells

The generalized equations of motion for both symmetric and asymmetric structures are obtained by incorporating the Rabi frequencies of interacting fields in the respective system in the  $3 \times 3$  interaction Hamiltonian matrix and applying

the slowly varying wave approximation. The off-diagonal elements of the density matrix satisfy,

$$\frac{\partial \rho_{jk}(t)}{\partial t} = -\{i\Delta_{jk} + \Gamma_{jk}\} \rho_{jk}(t) + i[\Omega_{jk} \{\rho_{kk}(t) - \rho_{jj}(t)\} + \Omega_{ji}\rho_{ik}(t) - \Omega_{ik}\rho_{ji}(t)] \quad (1)$$

where  $\Omega_{ji}$  is the Rabi frequency of the optical field tuned to the states  $|j\rangle$  and  $|i\rangle$ ,  $\Omega_c$  and  $\Omega_p$  are the Rabi frequencies of the coupling and the probe fields,  $\Gamma_{ji}$  is the dephasing rate between levels  $j$  and  $i$ , while the optical energies coupling the two states are defined as  $\hbar\Delta_{jk} = E_j - E_k - \hbar\omega_{jk}$ ,  $\rho(t)$  has the symmetric property,  $\rho(t)_{jk} = \rho(t)_{kj}^*$ .

The diagonal elements for a closed three-level quantum well system is represented by,

$$\frac{\partial \rho_{jj}(t)}{\partial t} = [(\partial\rho/\partial t)_{relax}]_{jj} + i[\Omega_{ji}\rho_{ij}(t) - \Omega_{ij}\rho_{ji}(t) + \Omega_{jk}\rho_{kj}(t) - \Omega_{kj}\rho_{jk}(t)] \quad (2)$$

where the relaxation Hamiltonian matrix element for the states  $|a\rangle$ ,  $|b\rangle$  and  $|c\rangle$  are considered respectively as,

$$\begin{aligned} [(\partial\rho/\partial t)_{relax}]_{aa} &= \rho_{bb}(t)\Gamma_{bb}, \\ [(\partial\rho/\partial t)_{relax}]_{bb} &= -\rho_{bb}(t)\Gamma_{bb} + \rho_{cc}(t)\Gamma_{cc}, \\ [(\partial\rho/\partial t)_{relax}]_{cc} &= -\rho_{cc}(t)\Gamma_{cc}, \end{aligned} \quad (3)$$

with  $T_{jj}$  ( $1/\Gamma_{jj}$ ) being the life time of the electrons at state  $|j\rangle$ . In Eq. (1) and (2) the indices  $i$ ,  $j$  and  $k$  refer to different subband states  $|a\rangle$ ,  $|b\rangle$  and  $|c\rangle$ . We have considered the system of three-level undoped quantum well to be completely closed satisfying the condition  $\rho_{aa} + \rho_{bb} + \rho_{cc} = 1$ . In symmetrical quantum wells, the electrons are assumed to decay from state  $|c\rangle$  to  $|b\rangle$  and from  $|b\rangle$  to  $|a\rangle$  with  $|a\rangle$  being assumed as the ground state ( $\Gamma_{aa} = 0$ ). In case of asymmetric structures there is an additional decay channel direct from state  $|c\rangle$  to  $|a\rangle$  for the electrons to decay back to the ground state without the intermediate conduction subband state  $|b\rangle$ . This leads to reduced interference between the interband and intersubband transitions that become significant in case of highly populated conduction subband states.

The analyses of the sum-frequency field generated due to the three wave interaction in an asymmetric well and its effect on the modulation of the probe field involves the transient evolution of the Maxwell - Bloch equations. The transient sum-frequency polarization is deduced from the wave equation under slowly varying envelope approximation as,

$$\frac{\partial F_s(t)}{\partial t} = -\frac{\omega_s}{\epsilon_0\epsilon_1} \text{Im}[P_s(t)], \quad (4)$$

where  $\epsilon_0$  is the free space dielectric constant, and  $\epsilon_1$  is the relative dielectric constant of the medium. The absorption of the signal due to internal losses has been neglected. We solve the set of six-coupled nonlinear optical Bloch equation obtainable from eqs.(1) and (2) by the fourth order Runge - Kutta method in case of the symmetrical quantum well structure whereas for the asymmetric structure the transient evolution of the generated sum-frequency signal is estimated by solving eq. (4) simultaneously along with eqs. (1) and (2).

The probe field dresses the initial population of the state  $|b\rangle$  and induces a finite polarization due to interband transitions. The electrons are coherently excited by the probe field prior to the switching of the coupling field, i.e., for the time  $t < 0$ , so that the initial conditions for eqs. (1) and (2) are,

$$\rho_{bb}(0) = \frac{2(\Omega_p^2 \Gamma_{ba}/\Gamma_{bb})}{\Delta_{ba}^2 + \Gamma_{ba}^2 + 4\Omega_p^2 \Gamma_{ba}/\Gamma_{bb}}, \quad (5a)$$

$$\rho_{aa}(0) = 1 - \rho_{bb}(0) - \rho_{cc}(0), \quad (5b)$$

$$\rho_{ba}(0) = \frac{\Omega_p(\Delta_{ba} + i\Gamma_{ba})\{1 - \rho_{bb}(0)\}}{\Delta_{ba}^2 + \Gamma_{ba}^2 + 4\Omega_p^2 \Gamma_{ba}/\Gamma_{bb}}, \quad (5c)$$

$$\rho_{cc}(0) = \rho_{cb}(0) = \rho_{ca}(0) = 0. \quad (5d)$$

The band-filling factor  $\{1 - \rho_{bb}(0)\}$  arising due to the accumulation of carriers in the first conduction subband state  $|b\rangle$  reduces the availability of vacant sites and reduces absorption in case of strong probe field. This factor has therefore been introduced in the steady state transition probability of the interband density matrix element in (5c).

The electronic polarization  $P$  arising from an interband probing field and an intersubband coupling field in the presence of the induced sum-frequency field is,

$$P(t) = \epsilon_0 \chi(t) [F_p(t) + F_c(t) + F_s(t)] \approx [\mu \rho(t) + \rho(t) \mu], \quad (6)$$

where  $\chi(t)$  is the complex susceptibility. The linear susceptibility  $\chi_{ba}(t)$  of the medium, at the probe frequency can be related to the absorption coefficient  $\alpha$  by its imaginary part  $\text{Im}[\chi_{ba}(t)]$  by assuming that the amplitude and polarization due to the probe field vary slowly over a wavelength or over a period. However the slowly varying envelope approximation approaches the limits of its validity for the case of femtosecond pulses with optical frequencies. It is observed that in case of a resonant coupling and probe field, when  $\Omega_c \gg \Gamma_{ba}$ , the dressed susceptibility  $\chi_{ba}(t)$  is described by a damped harmonic oscillator with initial amplitude  $\rho_{ba}(0)$ .

The physical characteristics of the modulation of the probe light due to the coupling pulses are analyzed with parameters appropriate for an actual  $\text{In}_{0.53}\text{Ga}_{0.47}\text{As}/\text{AlAs}$  quantum well grown over an InP substrate with a well width of 130 Å. The asymmetry in a symmetrical quantum well is induced by the application of a dc electric field of 30 kV/cm perpendicular to the layers. The steady-state perturbation theory predicts very strong quadratic interband sum-frequency nonlinearity with a negative sign at resonant frequencies [2]. Our investigation reveals that the response time of the material such as population lifetime and dephasing time sets a limit to the length of the coupling light pulses which might be employed for the efficient modulation of the probe light. The relative ratios of the coupling pulse width to the intersubband lifetime  $T_{cc}$  is one of the most significant parameters governing the transient modulation characteristics of the IB-resonant probe light in the presence of a coupling field. This is due to reason that for comparatively long  $T_{bb}$ ,  $T_{cc}$  determines the duration for which the carriers excited by the coupling field from the conduction subband state  $|b\rangle$  is retained at state  $|c\rangle$  thereby affecting the temporal absorption of probe light. The carrier density of the well does not effect the modulation process unless the Rabi frequency of the probe light is exceedingly lower than  $T_{ba}^{-1}$ . It is found that

the sum-frequency response depends on the initial dressing of the intersubband states [8]. The magnitude of the sum-frequency oscillation increases with the probe field strength as the population build up due to the pumping of carriers increases the electrons available at the state  $|c\rangle$  in the presence of intersubband resonant field and which subsequently decays back directly the ground state. The increase in probe field also induces a change in phase of the generated sum-frequency oscillations.

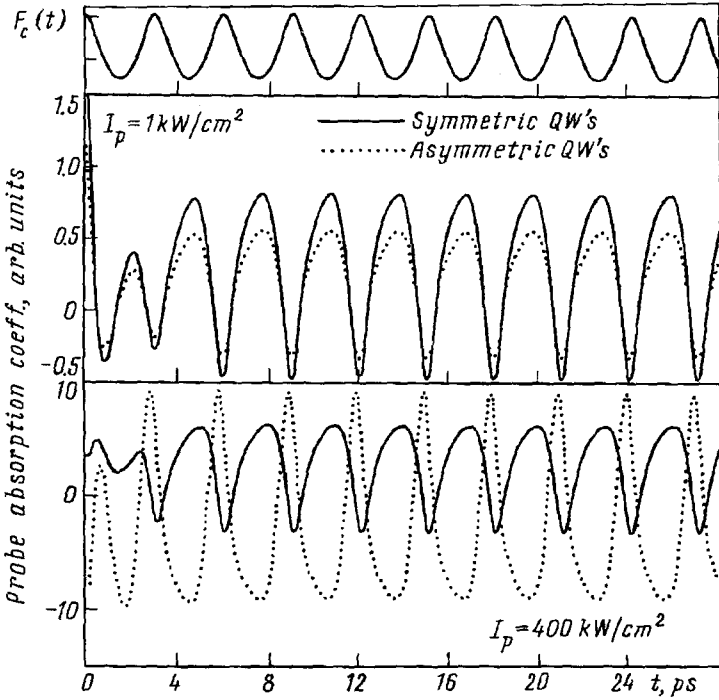


Fig.2. Comparison of transient interband probe absorption in symmetric and asymmetric wells for  $\tau_d > T_{ba}, T_{cc}$ ; with coupling intensity  $I_c = 1.0 \text{ MW/cm}^2$ ,  $\tau_d = 3.0 \text{ ps}$ ,  $\tau_p = 1.0 \text{ ps}$ ,  $T_{cc} = T_{ba} = 1.0 \text{ ps}$ ,  $T_{bb} = 1.0 \text{ ns}$ , (a)  $I_p = 1 \text{ kW/cm}^2$ ; (b)  $I_p = 400 \text{ kW/cm}^2$

In Fig.2 we have compared the modulation process in symmetric and asymmetric structures induced by a train of symmetric pulses with their widths ( $\tau_p$ ) and repetition rates ( $\tau_d$ ) larger than the phenomenological intersubband lifetime ( $T_{cc}$ ) and interband relaxation rates ( $T_{ba}$ ). The transient variation of the optical field amplitude of the coupling pulse train is depicted at the top of Fig.2 and 3. The interband probe absorption coefficient is alternatively positive and negative, implying that absorption and emission occur simultaneously. It has been noticed that in case of the modulation of weak probe fields when the nonlinearity due to the three-wave mixing is weaker the transient oscillatory features of the interband absorption are very similar to that in symmetric structures (Fig.2a). There is actually a slight reduction in the probe oscillations due to reduced interband transition dipole moment ( $\mu_{ba}$ ) owing to the asymmetry [9]. In the presence of stronger probe fields due to the dressing of the conduction subband states the initial population of electrons in the conduction subband state  $|b\rangle$  increases. This

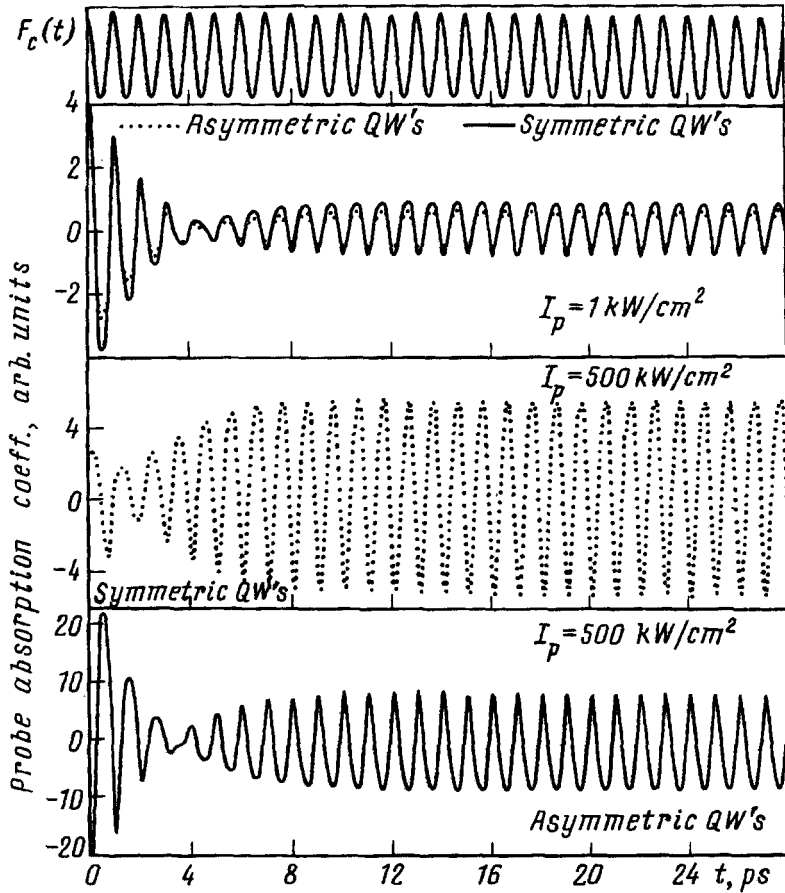


Fig.3. Comparison of transient interband probe absorption in symmetric and asymmetric wells or  $\tau_d < T_{ba}$ ,  $T_{cc}$  with coupling intensity  $I_c = 1.0 \text{ MW/cm}^2$ ,  $\tau_d = 1.0 \text{ ps}$ ,  $\tau_p = 400 \text{ fs}$ ,  $T_{cc} = 1.0 \text{ ps}$ ,  $T_{ba} = 3.0 \text{ ps}$ ,  $T_{bb} = 1.0 \text{ ns}$ , (a)  $I_p = 1 \text{ kW/cm}^2$ ; (b)  $I_p = 500 \text{ kW/cm}^2$  without sum-frequency field (c)  $I_p = 500 \text{ kW/cm}^2$  in presence of sum-frequency field

results in a population inversion between the interband states that leads to stronger oscillatory transient sum-frequency signal arising from the direct transition between the states  $|c\rangle$  and  $|a\rangle$ . The strength of the induced probe oscillations at higher  $F_p$  is also considerably enhanced in case of the asymmetric quantum wells as the transfer of energy from the nonlinear sum-frequency wave oscillates between the two interband fields. (Fig.2b). It can also be observed that in asymmetric wells in the presence of an appreciably strong sum-frequency field there is a change in the phase of the induced oscillations along with the input probe field strength. Thus an increase in the probe power level induces a change in the phase of the modulated oscillations under the influence of the nonlinear sum-frequency signal field.

In Fig.3 the effect of repetitive excitation of the coupling light on the probe modulation is shown for pulse delay time  $\tau_d$  shorter than interband dephasing time scale  $T_{ba}$ . As observed in Fig.2a at weaker probe field strengths the transient

response is similar for both symmetric and asymmetric quantum wells. In this case, during time scales lower than  $T_{ba}$ , the polarization left in the medium by the first pulse interacts with the second pulse resulting in the interaction of two pulses separated in time. In case of the modulation of a weak probe field it has been observed that due to a destructive interference effect suppressing the population build up despite repetitive pulse excitation the transient oscillatory behavior of the probe field is reduced. Therefore the switching of the coupling field with short delay times  $\tau_d$  is less effective on the induced interband polarization due to the destructive interference effect that reduces the oscillatory absorption characteristics of the weak probe light within time scales shorter or comparable to  $T_{ba}$  (Fig.3a). It can be further observed from Fig.3b that in symmetric structures if the probe field is appreciably stronger then at time scales larger than  $T_{ba}$  the transient oscillations build up proportionately to the magnitude of the Rabi frequency after the initial suppression of the induced oscillations during time scales  $t < T_{ba}$ . This characteristic is found to be reversed in the presence of the sum-frequency field in asymmetric wells (Fig.3c). The nonlinear sum-frequency field quenches the build up in the probe oscillation observed in Fig.3b at time scales longer than  $T_{ba}$  after the decay of the stronger initial oscillations during  $t < T_{ba}$ .

We have theoretically analyzed the influence of transient sum-frequency signal on the interband-resonant probe light modulation by a train of intersubband resonant light in an undoped quantum well. The presence of an additional sum-frequency signal that is adequately strong is expected to enhance the induced oscillations of the interband probe light. The magnitude of the instantaneous response of the medium at the sum-frequency depends on the extent of the population inversion between states  $|b\rangle$  and  $|a\rangle$  which can be controlled by the probe light. The most efficient modulation can be achieved in the presence of strong probe and coupling light fields. In case of modulation of ultrashort light pulses a stable modulation characteristic is achieved at time scales longer than the interband dephasing time ( $T_{ba}$ ) of the system. Possible applications of the proposed model include the realization of picosecond lasers in the visible and near-infrared regime.

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