Pump-probe measurement of ultrafast all-optical modulation based on intersubband transition in *n*-doped quantum wells

T. Asano,^{a)} M. Tamura, S. Yoshizawa, and S. Noda

Department of Electrical Science and Engineering, Kyoto University, Kyoto 606-8317, Japan

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Modulation of interband-resonant light (~800 nm) by intersubband-resonant light (5–7 μ m) was investigated in *n*-doped AlGaAs/GaAs multiple quantum wells by a two-color femtosecond pump-probe technique. Modulation with a recovery time of ~1 ps is observed in a plainer-type modulation device at room temperature. The modulation of interband absorption coefficient is ~1000 cm⁻¹ when the energy density of the intersubband light pulse is ~4 fJ/ μ m². The modulation efficiency indicates that 99% modulation can be achieved with a control pulse energy of ~1 pJ when a conventional waveguide-type device structure is utilized. The mechanism which determines the modulation speed is discussed in terms of carrier relaxation process. It is shown that the modulation speed is mainly determined by the inter- and intrasubband relaxation times, where the latter is influenced by hot phonon effects. © 2000 American Institute of Physics. [S0003-6951(00)02727-3]

The intersubband transition (ISB-T) in quantum well (QW) structures has interesting properties such as a large transition dipole moment in the midinfrared range¹ and an ultrafast energy relaxation time due to longitudinal optical (LO) phonon scattering (\sim picoseconds).^{2,3} The ultrafast relaxation process of the ISB-T is important for basic physics as well as for the development of high-speed optical devices. The authors previously proposed an ultrafast all-optical modulation scheme that utilizes the ISB-T.⁴ This scheme uses three subbands formed in a n-doped QW (Fig. 1): the first valence subband (VB1) and the first and second conduction subbands (CB1 and CB2). Interband (IB) resonant light absorption (VB1 \rightarrow CB1) can be increased by exciting the electrons from CB1 to CB2 using ISB-resonant light. The increased absorption decreases as the electrons relax from CB2 to CB1, and the relaxation time appears to be as fast as \sim 1 ps. Thus, it is thought that an IB-resonant light signal can be modulated using an ISB-resonant light signal on the order of picoseconds.

Theoretical and experimental investigations have revealed the static (not ultrafast) characteristics of the modulation.^{4–7} However, the modulation speed was not determined in the previous reports. In this letter, we report femtosecond time-resolved measurement results for the all-optical modulation. A planar-type GaAs/AlGaAs QW modulation device was fabricated and investigated using a two-color pump-probe technique. The modulation speed is shown to be ~1.3 ps. The relaxation process of the ISB-excited electrons is also discussed.

The modulation device investigated consists of 150 periods (N_{well}) of GaAs(59 Å)/Al_{0.35}Ga_{0.65}As (150 Å) multiple QWs grown on a semi-insulating GaAs substrate by molecular beam epitaxy. The barrier layer was selectively doped at a Si density of 1.5×10^{18} cm⁻³. A 0.5- μ m-thick Al_{0.4}Ga_{0.6}As etch stopping layer was grown between the QW layer and the

GaAs substrate. The device was fabricated by fixing the sample on a thin transparent plate, and the GaAs substrate was removed by selective wet etching in order to make the device transparent to IB-resonant light. The midinfrared absorption spectrum of the QWs was measured in the conventional Brewster angle configuration by Fourier-transform infrared spectroscopy (FTIR) [Fig. 3(b)]. An optical absorption peak due to the ISB-T was observed at 7.2 μ m (172 meV). The absorption peak was ~19%, and the full width at half maximum (FWHM) was ~20 meV. The near-infrared absorption spectrum of the sample was also measured and the IB absorption edge (VB1 \rightarrow CB1) was found to be about 830–840 nm.

Time-resolved modulation experiments were carried out using the two-color pump-probe method, in which the device is pumped by a midinfrared (4–9 μ m) control light pulse and is probed by a near-infrared (~800 nm) signal light pulse. The pump pulse was generated by optical parametric amplification and differential frequency mixing using an 800 nm mode-locked Ti–sapphire laser pulse. The residual of the 800 nm light pulse was used for the probe. The temporal and spectral width of the pump and probe pulses were ~120 fs and ~20 meV, respectively. The pump beam was incident to



FIG. 1. Schematic diagram of the all-optical-modulation scheme in a n-doped QW (a) without and (b) with control light.

19

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FIG. 2. Example of time-resolved all-optical-modulation characteristics. Inset is a logarithmic plot of the modulation depth.

the sample at the Brewster angle in order to ensure sufficient interaction with the ISB dipole moment and also to suppress multiple reflections inside the sample. The incident angle for the probe beam was almost the same as that of the pump beam. The effective energy density of the control pulse (J_c), which is actually absorbed by the QWs, was varied from ~2 to ~140 fJ/µm². A sufficiently small energy density of <~5 fJ/µm² was used for the signal pulse so as not to supply too many electrons from VB1 to CB1.

Figure 2 shows the temporal trace of the change in the transmittance of the probe (signal) pulse induced by the pump (control) pulse (i.e., the modulation profile), where J_c was ~4 fJ/ μ m² and the center wavelength of the control pulse (λ_c) was tuned to the ISB-resonance (7.2 μ m). The rapid decrease in the transmittance and its ultrafast recovery can be clearly seen in the figure. The recovery time of the modulation (measured as FWHM of the modulation profile) is as short as 1.3 ps. We also carried out the measurements while gradually detuning λ_c from the ISB resonance. Figure 3 shows the peak of the modulation depth measured as a function of λ_c together with the ISB absorption spectrum of the QWs. The correlation between the modulation and absorption spectra clearly demonstrates that the observed modulation is due to the ISB-T.

The peak modulation depth observed in the first measurement (Fig. 2) corresponds to the increase in the absorption coefficient of as much as $\sim 1000 \text{ cm}^{-1}$, where the energy density of the control pulse was $\sim 4 \text{ fJ}/\mu\text{m}^2$. It is estimated that $\sim 99\%$ modulation of the signal light can be



FIG. 4. Relaxation curve of ISB absorption saturation measured by onecolor pump-probe method. The sample was pumped and also probed by ISB-resonant light pulses (\sim 7.2 μ m).

obtained with a control pulse energy of ~ 1 pJ when a conventional waveguide-type device configuration, which can enhance the interaction between QWs and light, is used instead of the surface incident configuration used here. The relatively high modulation efficiency is due to the fact that the transition dipole moment of the ISB-T in the midinfrared range is very large (~ 17 eÅ in this QW) and the near-infrared photon (~ 1.5 eV) is controlled by the midinfrared photon (~ 0.17 eV).

The inset in Fig. 2 shows a logarithmic plot of the modulation profile. The modulation decays with a short relaxation time of $\sim 1.1 \text{ ps} (T_a)$ soon after the ISB-excitation (delay time $0 \sim 1$ ps), and subsequently the relaxation time becomes longer (~3 ps: T_b). Furthermore, we measured the recovery time of ISB absorption saturation in the same QWs, where the QWs were pumped and probed by the ISB-resonant light pulses (Fig. 4). It can be seen in Fig. 4 that the ISB absorption recovery time (T_{isb}) is 0.6–0.7 ps. The discrepancy between T_a and T_{isb} can be explained in terms of the relaxation process. In the latter case, the pump-induced absorption saturation recovers when the excited electrons are scattered from CB2 to the higher energy part of CB1 since the two subbands are almost parallel [Fig. 5(a)] However, in the former case, the electrons must relax from the higher energy part of CB1 to the bottom of CB1 (intrasubband energy relaxation) in order to reduce the IB absorption near the band edge [Fig. 5(b)]. The intrasubband energy relaxation process is expected to take an additional 0.3–0.4 ps since the ISB energy spacing ($E_{isb} \sim 172 \text{ meV}$) is about 4–5 times that of the LO



FIG. 3. (a) Peak modulation depth measured as a function of the control light wavelength (data for the longer wavelength side could not be measured due to limitations in the light source), and (b) infrared absorption spectrum of the sample measured by FTIR.





FIG. 5. Schematic diagram showing the electron relaxation process for recovery of ISB absorption saturation (a) and the proposed modulation (b).

phonon energy and each LO phonon emission takes ~0.1 ps.³ (Electron–electron scattering is much faster, but it is not a energy relaxation process). Therefore, the discrepancy between $T_{isb}(0.6-0.7 \text{ ps})$ and $T_a(\sim 1.1 \text{ ps})$ can be explained by the additional intrasubband energy relaxation time (0.3–0.4 ps). We assumed that the long relaxation time observed in the subsequent decay process ($T_b:\sim 3$ ps) is due to the reabsorption of hot phonons emitted during intrasubband relaxation.^{8,9} (The hot phonons emitted by the ISB scattering have no effect on intrasubband phonon scattering due to the different symmetry.) Then, we estimated the hot LO population (number of phonons per mode) that is generated by the complete intrasubband energy relaxation for the case in the present experiment. The total number of hot LO phonons (n_p) can be estimated from J_c by the equation¹⁰

$$n_p = \frac{J_c / E_{\rm isb}}{N_{\rm well}} (E_{\rm isb} / \hbar \,\omega_{\rm LO} - 1). \tag{1}$$

The number of LO phonon modes that contribute to the intrasubband relaxation (N_p) can now be estimated by the following equation:³

$$N_p = \frac{2q_{\parallel\min}^2}{\pi} N_\perp \,, \tag{2}$$

where $q_{\min} = \sqrt{2m^*(E_{isb} - \hbar \omega_{LO})}/\hbar - \sqrt{2m^*(E_{isb} - 2\hbar \omega_{LO})}/\hbar$ is the minimum in-plane momentum (q_{\parallel}) of the LO phonon which is emitted during the intrasubband relaxation process.³ In the equation, we cut off the phonon modes whose q_{\parallel} is greater than $3q_{\parallel \min}$ since the electron-LO phonon interaction strength is proportional to $1/q_{\parallel}^2$. N_{\perp} is the number of vertical modes of the LO phonon. We assumed that two vertical phonon modes (the first mode of a GaAs-wellconfined phonon and the symmetric mode of a AlAsinterface-trapped phonon) are the main contributors to the relaxation.³ The average hot phonon population (n_{av}) can be calculated from the equation $n_{av} = n_p / N_P$. By using these equations and conventional material parameters, n_{av} is estimated to be ~ 0.2 in the case of the experiment shown in Fig. 2, which is as large as the LO phonon population in thermal equilibrium at 300 K. Since the relaxation time of hot LO phonons is as long as 5-7 ps,^{11,12} it is possible that electron intraband relaxation becomes slower by an order of picoseconds as a result of the hot phonon reabsorption process. Moreover, in additional experiments, it was observed that T_b becomes longer from ~3 to ~7 ps as J_c increases from ~2 to ~140 fJ/ μ m². The results suggest that T_b is influenced by the effects of hot phonon reabsorption. Therefore, utilization of a waveguide-type configuration with a low energy density control pulse is preferable since hot phonon effects can be avoided.

In conclusion, we carried out time-resolved measurement of an all-optical modulation scheme that utilizes the ISB-T by means of a two-color pump probe technique. It was directly shown that ultrafast operation in the range of 1.3 ps (FWHM) is possible in this scheme using a relatively low control light pulse of 4 fJ/ μ m². The results indicate that the ISB-T is useful for realizing ultrafast low-energy all-optical modulation and switching.

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