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Wavelength selective photoexcitation of picosecond acoustic-phonon pulses in a triple GaAs/Al_{0.3}Ga_{0.7}As quantum well structure

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Abstract

Ultrashort coherent phonon-pulse generation and detection is investigated in a GaAs–Al_{0.3}Ga_{0.7}As quantum well structure containing three wells of different widths using an optical pump and probe method. The pump photon energy is tuned to the region of the $hh1-e1$ transition energies of the wells and the probe photon energy is chosen for detection of the phonon pulses that reach the sample surface. By studying the dependence of the probe reflectance change on the pump photon energy, we demonstrate the possibility of wavelength-selective excitation of picosecond acoustic-phonon pulses in quantum wells. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

The recent development of ultrashort-pulse mode-locked lasers has allowed the excitation and detection of high-frequency acoustic-phonon pulses in solids. This technique, known as laser picosecond acoustics, relies on the excitation of acoustic-phonon pulses with sub-picosecond pump laser pulses and their detection with appropriately delayed probe pulses [1]. It has been applied to a wide variety of metallic and semiconductor films and nanostructures.

Application to semiconductor nanostructures is interesting for elucidating the basic physics of phonon generation and propagation in confined quantum geometries. Quantum nanostructures offer a vast testing ground in this field for probing phonon generation mechanisms, hot electron relaxation and electron–phonon interactions. By exploiting the techniques of band engineering, well-defined and highly confined phonon generation regions can be realized. Such structures could serve as high-frequency THz phonon transducers to be used, for example, for phonon spectroscopic studies. Preliminary experiments involving the application of laser picosecond acoustics to GaAs/AlGaAs quantum wells have been reported [2,3]. It was shown that ultrashort

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acoustic-phonon pulses can be generated in buried quantum wells and detected at the sample surface. Quantum wells have a sensitive optical-wavelength tunable response owing to their excitonic resonances defined by accurately characterized wave functions. We exploit this tunability in the present study by demonstrating how the phonon generation region can be optically selected for a triple quantum well structure.

2. Experiment and results

A GaAs/Al_{0.3}Ga_{0.7}As quantum well structure was prepared on a GaAs (100) substrate using metal organic vapor phase epitaxy (MOVPE) to have three buried GaAs quantum wells of different thicknesses (and hence different excitonic optical resonances). Details of the design are shown in Fig. 1. The calculated $hh1-e1$ transition energies at 300 K for isolated quantum wells with widths corresponding to the labels A, B, and C in Fig. 1 are 1.55, 1.48 and 1.61 eV, respectively. Photoluminescence measurements at room temperature show three distinct luminescence peaks at

around 1.51, 1.46 and 1.56 eV, that are reasonably close to the design transition energies of the three wells.

Infrared optical pump pulses of photon energy tunable in the range 1.46–1.57 eV (wavelength 840–790 nm), duration ~ 700 fs, and repetition rate 82 MHz from a mode-locked Ti:Sapphire laser are used to excite longitudinal acoustic-phonon pulses in the quantum wells. The pump photon energy range spans the $hh1-e1$ transition of the three wells, low enough for the light to be transmitted by the Al_{0.3}Ga_{0.7}As barrier layers. The pump light is focused onto the sample surface with a spot diameter ~ 20 μm , with an incident fluence ~ 0.3 mJ cm^{-2} per optical pulse. The phonon pulses excited inside the sample can be detected at the top surface through the change in complex reflectance arising from the photoelastic effect and from the surface displacement. A blue probe beam (with a penetration depth of < 10 nm) of delayed optical pulses of duration ~ 200 fs derived by doubling the pump photon energy is used for detection of the change in (complex) reflectance $\delta r/r = \rho + i\delta\phi$ in the near-surface region of the sample using a Sagnac interferometer [4]. The fluence of the probe pulses is ~ 0.03 mJ cm^{-2} and the spot diameter is ~ 10 μm .

Some representative results for the real (ρ) and imaginary ($\delta\phi$) parts of the optical reflectance as a function of delay time are shown in Fig. 2 for pump photon energies 1.57 and 1.49 eV. Features appearing at delay times > 230 ps correspond to the arrival at the surface of phonon pulses generated in the GaAs substrate. The vertical scales have been adjusted to make this substrate signal equal in all the graphs. The signals at ~ 100 ps correspond to the arrival at the surface of phonon pulses generated in the quantum wells. The overall duration for this quantum well signal is significantly longer for the 1.57 eV excitation than that for the 1.49 eV excitation. This is not unexpected considering the optical absorption characteristics of the wells: the lower pump energy (1.49 eV) should excite phonon pulses only at well B, while the higher pump energy (1.57 eV) should do so in all the wells. (Well B is the thickest and thus has the lowest $hh1-e1$ transition energy.)

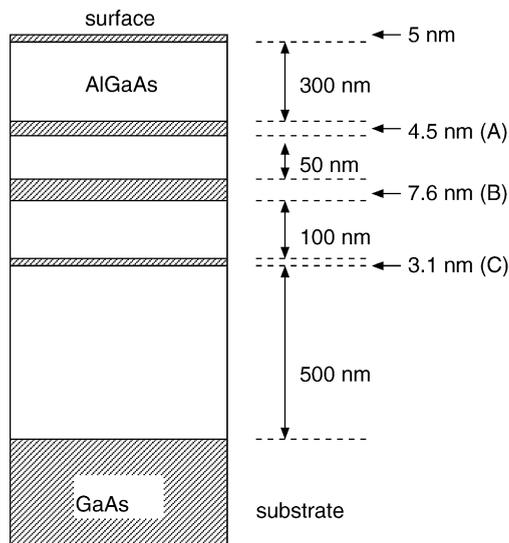


Fig. 1. Dimensions of the GaAs/Al_{0.3}Ga_{0.7}As quantum well structure.

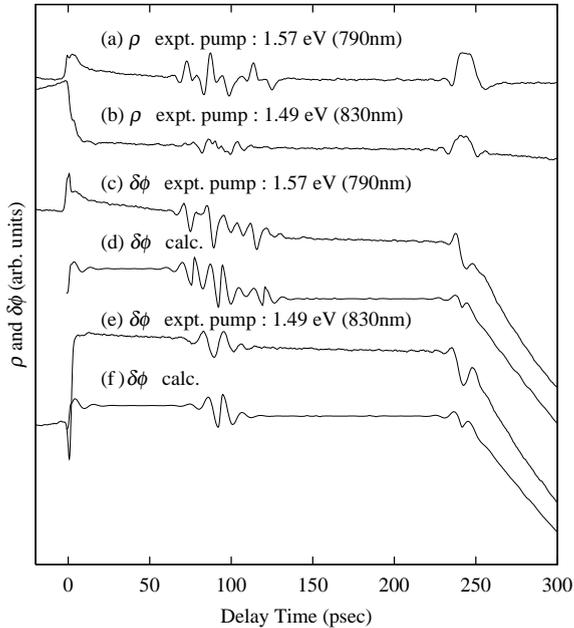


Fig. 2. Real (ρ , (a) and (b)) and imaginary ($\delta\phi$, (c)–(f)) parts of the reflectance change at pump photon energies 1.57 and 1.49 eV. The pump-energy dependence of the phonon generation region is clearly seen from the difference in signal shape at ~ 100 ps. Calculated results for $\delta\phi$ are shown in (d) and (e) for the cases of three-well excitation and single-well (B) excitation, respectively.

3. Discussion

In order to interpret the experimental data further, we have performed a simulation that takes into account the phonon generation, propagation, and detection processes. The absorbed photons set up an excited electron–hole (e–h) distribution and also produce a change in lattice temperature distribution governed by the relaxation of this excited e–h distribution. These e–h and temperature distributions lead to an initial stress mediated by the deformation potential and the thermal expansion coefficient, respectively. For semiconductors the deformation potential mechanism is often the dominant mechanism, and this is expected to be the case here [3,5–7]. We therefore assume that the spatial distribution of the initial stress is proportional to that of the initial excited e–h density, independent of the excess electron energy. The excited e–h density

depends on the optical constants of the wells and on the spatial profile of the appropriate wave function(s). However, for simplicity, we assume here that this spatial profile is a top-hat function defined by the well width.

The phonon propagation is calculated from the one-dimensional elastic wave equation, using literature values of the longitudinal sound velocities 4.73 and 4.95 nm ps⁻¹ for the GaAs and Al_{0.3}Ga_{0.7}As layers, respectively [8]. The multiple acoustic reflection problem is treated using a knowledge of the acoustic impedance mismatch between the GaAs and Al_{0.3}Ga_{0.7}As layers. (For example, the acoustic reflectance for strain at such an interface is ~ 0.02 .) Frequency-dependent ultrasonic attenuation is not included here for simplicity.

The travelling acoustic-phonon pulses modulate the optical properties of the sample inhomogeneously, and a general treatment of this optical modulation is possible [9]. Here, the inhomogeneous perturbation occurs near the sample surface within the probe light penetration depth. Fig. 2(d) and (f) show fitted results for $\delta\phi(t)$ for the case in which all three quantum wells are excited and for the case in which only one quantum well (B) is excited, respectively. The overall agreement with experiment is good.

4. Conclusions

In conclusion, we have studied the acoustic-phonon generation and detection in a GaAs/Al_{0.3}Ga_{0.7}As quantum well structure using laser picosecond acoustics. The excitation-wavelength dependence of the echo shapes conclusively demonstrates that we have achieved wavelength-selective phonon excitation in the quantum wells. This ease of optoacoustic tuning should lead to interesting applications in high-frequency phonon spectroscopy.

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