1.26 $\mu$m intersubband transitions in In$_{0.3}$Ga$_{0.7}$As/AlAs quantum wells

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Abstract

We observed room-temperature intersubband transitions at 1.26 microns in n-doped type-II In$_{0.3}$Ga$_{0.7}$As/AlAs strained quantum wells. An improved tight-binding model was used to optimize the structure parameters in order to obtain the shortest wavelength intersubband transition ever achieved in a semiconductor system. The corresponding transitions occur between the first confined electronic levels of the well following mid-infrared optical pumping of electrons from the barrier X-valley into the well ground state.

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Intersubband optical transitions (ISTs) between quantized electronic levels in semiconductor quantum wells (QWs) are at the basis of the realization of novel mid-infrared semiconductor detectors and lasers. In recent years much attention was given to the implementation of systems exhibiting ISTs with shorter wavelengths extending to the near infrared. In fact, thanks to the ultrafast carrier-relaxation processes characteristic of intersubband scattering such high-energy ISTs can find many important applications for high-speed device operation at short wavelengths and for ultrafast all-optical modulation schemes that would require similar wavelengths for both interband and intersubband transitions. The achievement of short-wavelength intersubband transitions is basically connected to the choice of suitable material combinations yielding adequately large conduction-band offsets. In particular, systems composed by narrow In$_x$Ga$_{1-x}$As/AlAs strained QWs were extensively studied and yielded wavelengths as short as 1.59$\mu$m and 1.55$\mu$m, thanks to the large In$_x$Ga$_{1-x}$As/AlAs conduction-band offset and large tunability of the IST energies. Picosecond intersubband relaxation in this material system for the case of ISTs at around 1.8$\mu$m was also recently demonstrated. The use of InGaAs/AlAsSb compounds has shortened these wavelengths down to 1.45$\mu$m which represents the highest-energy IST reported so far. Additionally, GaN/AlGaN narrow QWs have been studied very recently and displayed intersubband transition at 1.77$\mu$m. This latter material combination may offer important avenues for the achievement of very short-wavelength ISTs.

In our recent work we demonstrated that n-doped In$_x$Ga$_{1-x}$As/AlAs heterostructures allow a large tunability of IST energies and can yield intersubband wavelengths well below 1.5$\mu$m. We also showed, however, that quantum-size effects associated with well thickness and indium mole fraction values can induce a type I$\rightarrow$type II transition as the QW width is decreased. This happens when the quantum-confined $\Gamma$ conduction-band minimum localized in the In$_x$Ga$_{1-x}$As well is pushed above the bulk AlAs X conduction-band minimum, leading to an indirect band-gap configuration. In this case, at equilibrium, electrons populate the X valley in the barrier. Intersubband absorption thus occurs in the mid-infrared and is associated with indirect electronic transitions from the X (barrier) to the $\Gamma$ (well) states.
Observation of short-wavelength direct intersubband transitions below 1.5 \( \mu m \) is hindered by the lack of electrons in the lowest \( \Gamma \)-like subband of the QW.

In this letter we shall focus on n-doped type-II In\(_{x}\)Ga\(_{1-x}\)As/AlAs multiple QW structures with narrow well widths (few monolayers) and we shall demonstrate that it is possible to photo-activate the very short-wavelength direct intersubband absorption associated to confined \( \Gamma \) states in the QWs by photo-pumping electrons out of the barrier X levels and into the lowest \( \Gamma \)-like state of the wells. The approach used here allowed us to circumvent limitations associated with the type-II character of these ultrathin QWs. As a result, we obtained room-temperature IST at 1.26 microns (0.98 eV) in a 5-monolayer(ML)-thick In\(_{0.3}\)Ga\(_{0.7}\)As/AlAs multiple-QW structure (1 ML \( \approx \) 3 Å). This observation sets the new record for the shortest-wavelength IST achieved in a semiconductor system.

In order to identify the optimal In\(_{x}\)Ga\(_{1-x}\)As/AlAs configuration for short-wavelength ISTs with large dipole moment, band-structure calculations were performed within the tight-binding (TB) approximation using a 40-band empirical \( sp^3d^5s^* \) nearest-neighbor model that includes spin-orbit coupling\(^1\). The usefulness of the present TB model was recently demonstrated in the calculation of the optical properties of several III-V semiconductor quantum wells and superlattices\(^2\) including the InGaAs/AlAs heterostructure system here of interest\(^3\).

Figure 1 (left panel) shows the calculated squared dipole matrix element \( E_P \) (in eV) of the predominant IST between the \( \Gamma \)-like conduction-band states, named c1 and c2, as a function of alloy composition and well thickness (\( E_P = \frac{2}{m_o} \langle c1 \mid P_z \mid c2 \rangle^2 \), where \( m_o \) is the free electron mass). The corresponding intersubband transition energies are displayed in the right panel of Fig. 1. These calculations suggest that useful, very short-wavelength intersubband transitions can indeed be achieved in this material system with ultrathin QWs. In particular, samples with well width of 5 MLs emerge as most promising candidates for short-wavelength intersubband absorption. In fact, 4-ML-wide QWs that present even larger IST energies do not yield an efficient localization of the c2 \( \Gamma \)-like state in the InGaAs QW region, which reflects in the low intersubband dipole moments reported in Fig. 1 (left panel). Setting the
QW thickness at 5 ML, however, demands much care in the choice of the alloy composition. At large In content (x > 0.6), these structures exceed the critical thickness as calculated following Matthews and Blakeslee’s mechanical-equilibrium model. From our calculations, the best In concentration value for the realization of short-wavelength IST is estimated at around x = 0.3. It must be noted that this well width (5ML) and In concentration lead to a type-II configuration and intersubband absorption in the mid-infrared as pointed out above. We shall demonstrate, however, that photoinduced occupation of the confined Γ-like subband of the QW enables the observation of the c1-c2 intersubband absorption.

The sample used in this study was grown by molecular beam epitaxy on a GaAs(001) substrate and consists of a 0.5 µm GaAs buffer layer followed by 30 In$_{0.3}$Ga$_{0.7}$As QWs 5-ML wide, grown at 540°C with a 30 s growth interruption at each interface, separated by 108-Å-thick (36ML) AlAs barriers. Wells were Si doped to 1.4×10$^{19}$ cm$^{-3}$. The growth was concluded with a 10-nm-thick GaAs cap layer. Well and barrier thickness were verified by transmission electron microscopy. The sample was fabricated in a multipass waveguide geometry with 45° polished mirror facets and placed in a Fourier Transform Infrared spectrometer (FTIR) for the mid-infrared absorption measurements.

Figure 2 shows the room-temperature mid-infrared absorption of the sample. The spectrum was acquired with z-polarized incident light (perpendicular to the QW plane) and normalized with respect to the in-plane polarized transmission signal. The absorption peak is centered at 98 meV (∼ 12 µm) and corresponds to the X-Γ indirect transition. This transition is induced by Γ-X intervalley mixing and is negligible for in-plane polarization owing to the reduced overlap between the envelope functions of the two states (see inset of Fig. 2). It must be noted that this selection rule is peculiar to our system; in short-period type II superlattices, for instance, where the X state is quantum-confined and therefore the wavefunction overlap is larger, the X-Γ intersubband transition is basically polarization insensitive.

The energy-level diagram of the heterostructure as derived within our TB approximation is shown in the inset of Fig. 3. The c1-c2 intersubband transition is calculated at 0.94 eV
In order to observe the short wavelength c1-c2 IST we used the radiation from a CO$_2$ laser to pump electrons out of the X level to the c1 Γ-like ground state in the well. During the experiment the sample was illuminated on the same spot (a few millimeters wide) with both the CO$_2$ laser and a halogen lamp for the transmission measurements. The CO$_2$ laser radiation was tuned at 110 meV (within the X-Γ absorption peak shown in Fig. 2) and focused on the sample surface with an intensity of \( \approx 20 \text{ W/cm}^2 \). This high power density was motivated by the low oscillator strength of the indirect intersubband transition \( f=0.001 \) compared with \( f\approx3.3 \) for the c1-c2 transition. The transmission signal was then dispersed by a 32-cm-long monochromator and detected by a Ge detector and conventional lock-in techniques.

Figure 3 shows the photo-activated intersubband absorption at room temperature. The peak is centered at 0.98 eV, \( \approx 1.26 \mu\text{m} \), in good agreement with the TB calculation results (see Fig. 1) and has a full width at half maximum of 60 meV which is largely due to well-width and alloy fluctuations. Data plotted were obtained by careful normalization to the transmission background signals obtained without concurrent CO$_2$ excitation. In order to rule out the influence of local heating effects on the observed near-infrared absorption peak we checked the impact of a different normalization procedure. To this end, we acquired transmission data by using as normalization background signal the transmitted intensity under concurrent laser irradiation at 135 meV with an intensity of \( \approx 20 \text{ W/cm}^2 \). (The excitation frequency lays out of the absorption peak shown in Fig. 2.) The same absorption peak was observed, confirming unambiguously that photo-pumping across the mid-infrared X-Γ transition is the dominant mechanism that yields electrons in the well. We also found that the c1-c2 absorption peak increased with excitation power further highlighting the photoassisted nature of the observed signal.

In conclusion, we have reported the experimental observation of room-temperature intersubband transition at 1.26 microns in type-II In$_{0.3}$Ga$_{0.7}$As/AlAs quantum wells. For this experiment, electrons were photo-pumped from the X valley located in the AlAs barrier.
to the lowest confined Γ-like state in the well by means of CO$_2$ laser irradiation. Further engineering of InGaAs/AlGaAs heterostructures that will take advantage of quantum confinement of X-valley levels in the barrier will open the way to the exploitation of very short wavelength IST in optoelectronic devices even without the need for MIR-photoexcitation.

This work was supported in part by the Consiglio Nazionale delle Ricerche (CNR) within the CNR-Scuola Normale Superiore framework agreement and by MURST. Two of the authors (CPG and BHM) acknowledge financial support from the European Commission under the ERASMUS and TMR programs, respectively. One of the authors (ADN) acknowledges IFAM-CNR for financial support. We thank A. Parisini and P.G. Merli for the transmission electron microscopy analysis.
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FIGURES

FIG. 1. Lowest dominant intersubband transitions between Γ-like conduction states (c1,c2) as a function of well thickness (in monolayers, ML) and alloy composition. Left Panel: Calculated dipole matrix element squared ($E_P$ in eV). Right Panel: Calculated intersubband transition energy ($\Delta E$). The oscillator strength (f) for each transition is given by $f=E_P/\Delta E$.

FIG. 2. Room-temperature mid-infrared intersubband absorption of 5-monolayer In$_{0.3}$Ga$_{0.7}$As quantum wells confined by AlAs barriers. The inset shows the polarization dependence of the intersubband absorption peak. Zero degrees corresponds to incident light polarized along the growth (z) direction.

FIG. 3. Room-temperature near-infrared intersubband absorption of 5-monolayer In$_{0.3}$Ga$_{0.7}$As quantum wells confined by AlAs barriers during CO$_2$ laser irradiation at 110 meV and $\approx 20$ W/cm$^2$ incident intensity. Each data point corresponds to 30-s integration time. Transmission signals were normalized to signals obtained without CO$_2$ laser irradiation. The inset displays the calculated band alignment of the heterostructure. The calculated c1-c2 intersubband transition energy is 0.94 eV. The dashed line indicates the CO$_2$-induced electron photopumping across the X-Γ transition.
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