Delayed luminescence induced by intersubband optical excitation in a charge transfer double quantum well structure

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Photoluminescence processes in a novel GaAs double quantum well (DQW) structure were studied and found to be controlled by a 10 μm mid-infrared (MIR) light. In this DQW structure, photogenerated electron-hole pairs are normally separated into different wells and their radiative recombination is inhibited. However, when a MIR light pulse is supplied to induce an electron intersubband transition, electrons are efficiently transferred to the hole-rich well, resulting in a significant enhancement of luminescence. By time shifting the MIR pulse with respect to the light pulse for interband excitation, we demonstrated the generation of a delayed photoluminescence. Device potentials of this MIR to near-infrared conversion are discussed. © *1997 American Institute of Physics.* [S0003-6951(9)01409-5]

Intersubband transitions (ISBT) in quantum wells have recently attracted much attention in the world of mid-infrared (MIR) physics. Efficient intersubband MIR detectors[1] and lasers[2] have been realized. In double quantum wells, ISBT induces a spatial transfer of electrons from one well to the other, leading to a large optical rectification in doped samples,[3] and to a modulation of photoluminescence (PL) intensity in nondoped samples.[4,5]

Here we present for the first time, to the best of our knowledge, a novel phenomenon in a charge transfer double quantum well structure (CTDQW)[6] of Fig. 1, in which electron-hole recombination is induced by ISBT, affording a new scheme to control near-infrared (NIR) light emission by MIR light. In a CTDQW, photogenerated carriers are separated by an applied electric field, so that holes accumulate in the left-hand side quantum well (QW_L) and electrons in the right-hand side quantum well (QW_R). High external barriers (B_E) prevent the escape of carriers; a wide middle barrier (B_M) separates electrons and holes and enhances their lifetime. In this system, we investigated an ISBT process, by which electrons in QW_R are transferred to the hole-rich QW_L, resulting in a radiative recombination.

A CTDQW sample was designed in order to have one electron subband confined in QW_L (level L), another in QW_R (level R), and a third above B_M (level 3) whose wave function extends over both wells (cf. Fig. 1). Intersubband transition energy between levels R and 3 was designed to be 120 meV to resonate with CO_2 laser light. The sample was prepared by molecular beam epitaxy on a semi-insulated GaAs substrate. From the top of the sample, the composition is: a 10 nm GaAs cap layer, a 200 nm superlattice spacer layer of AlAs/GaAs (3 nm/0.7 nm), 16 periods of CTDQW, a 102 nm superlattice buffer layer of GaAs/AlAs (0.6 nm/4.5 nm), and a 50 nm heavily n-doped GaAs layer. Each period of CTDQW consists of an external barrier (B_E) containing four periods of AlAs/GaAs (3 nm/0.7 nm) superlattice, a 8 nm GaAs well (QW_L), a 25 nm $Al_{0.2}Ga_{0.8}As$ barrier (B_M) and a 6.5 nm GaAs well (QW_R). A Schottky contact was formed by depositing a 10 nm semitransparent Au layer on the front surface. An ohmic contact was made on the doped layer. A reverse bias voltage was applied normally to the well layers to separate the photogenerated electron-hole pairs. Using this 0.3-mm-thick wafer, a 2-mm-long multipass waveguide for MIR was produced by polishing two opposite ends at an angle of 45° (cf. inset of Fig. 4).

First, the sample was cooled at 10 K and a reverse bias $U=-1.5$ V was applied. Carriers were generated by supplying a 33-μs-long Ar^+ laser pulse of 120 mW/cm^2 through the semitransparent gold layer. The dotted line in the inset of Fig. 2 shows the PL spectrum measured during this excitation. By comparing with theoretically calculated quantum levels, the peak at $\lambda=783$ nm is ascribed to QW_L, and the peak at 771 nm to QW_R. Though the PL intensity from QW_R is much weaker than the PL of QW_L under reverse bias, it increases and becomes larger than the PL from QW_L in the forward bias region. This behavior is characteristic of CTDQW structures and denotes an efficient carrier separation.[7] As shown in Fig. 2, the PL intensity from QW_L increased gradually with time t during Ar^+ laser excitation. This increase results from the accumulation of spatially separated electrons and holes which screen the applied electric field, since the screening of the field reduces carrier separation, and enhances radiative recombinations.[6]

When the Ar^+ laser was turned off at $t=0$, PL signal quenched rapidly (point B), indicating that accumulated elec-

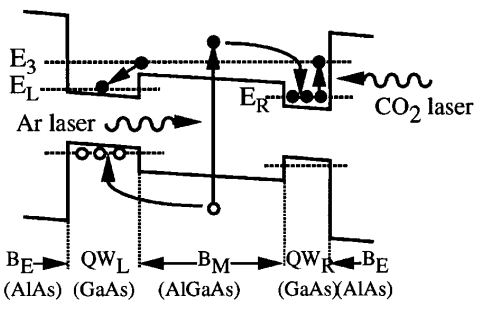

FIG. 1. A charge transfer double quantum well (CTDQW) structure under reverse bias. Photogenerated electron-hole pairs are spatially separated. Electrons from QW_R can be excited by intersubband transition to a subband above B_M, from where they can relax in QW_L.

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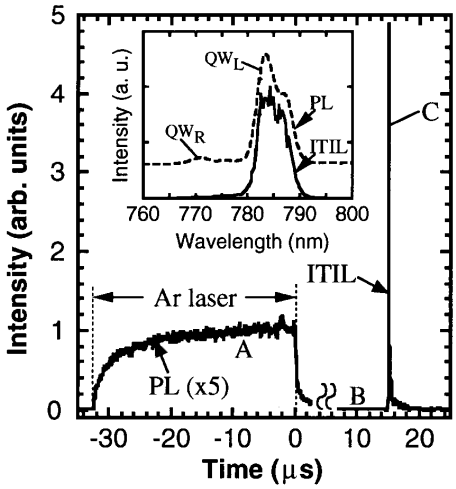

FIG. 2. The time variation of luminescence of QW_L measured at 10 K. First the sample is illuminated during 33 μs by an Ar^+ laser beam of 120 mW/cm^2. Then, a TM-polarized 100-ns-long CO_2 laser pulse of 70 kW/cm^2 and of photon energy $\hbar\omega_{CO_2}=120.68$ meV is applied 15 μs after the Ar^+ laser was turned off. A reverse bias of -1.5 V is applied. The inset shows the PL spectrum during Ar^+ laser excitation (broken line) and the spectrum of ITIL induced by CO_2 laser excitation (solid line). The spectra are normalized and shifted to facilitate the comparison.

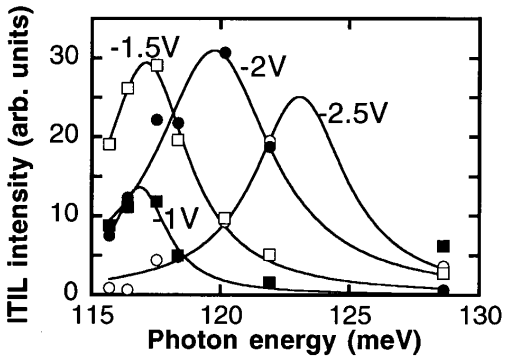

FIG. 3. The time integrated intensity of ITIL as a function of CO_2 laser photon energy measured under several bias voltages. A cw Ar^+ laser light of 5.4 mW/cm^2 was used for electron-hole pair generation, and CO_2 laser pulses of 40 kW/cm^2 for ITIL generation. The background PL by Ar^+ laser has been subtracted from the data. The symbols denote data points and the solid lines are Lorentzian fits.

trons and holes do not recombine radiatively. Some time Δt after the end of Ar^+ laser pulse, we illuminated the sample through one of the 45° end faces with a 100-ns-long TM-polarized CO_2 laser pulse of 70 kW/cm^2 and of photon energy $\hbar\omega_{CO_2}=120.68$ meV. We found that a very intense luminescence was emitted from QW_L, when the delay Δt was 15 μs (point C), as shown in Fig. 2. We call this luminescence the intersubband-transition-induced luminescence (ITIL), hereafter, for reasons mentioned below. Note that ITIL is about 20 times more intense than the preceding PL signal. When Ar^+ laser power was reduced, ITIL intensity decreased in proportion, showing that ITIL results from the recombination of carriers generated by Ar^+ laser. The spectrum of ITIL is shown by the solid line in the inset of Fig. 2. A peak appears at $\lambda=783$ nm, indicating ITIL results from the recombination of electron-hole pairs in QW_L. No luminescence from QW_R was observed. Since no electrons are initially in QW_L, we infer that MIR excitation induces a transfer of electrons from QW_R to QW_L. In the following, we study the mechanism of transfer.

Figure 3 shows the relation between ITIL intensity and the photon energy of CO_2 laser, for four different bias voltages. Here, a cw Ar^+ laser light of 5.4 mW/cm^2 was used for electron-hole pair generation, and a TM-polarized CO_2 laser pulse of 40 kW/cm^2 for ITIL generation. The background PL by Ar^+ laser was subtracted from the data. ITIL intensity peaks at a photon energy close to the designed subband spacing, implying that it is induced by an ISBT. When TE-polarized CO_2 laser light was used, ITIL intensity reduced to a fourth of its value, strongly suggesting an electron ISBT. In addition, the energy of the peak response moves sensitively to higher energy as the bias becomes more negative. This indicates that the ISBT occurs between levels R and 3, and not between levels L and 3, since the theory predicts that the spacing between levels 3 and R increases, whereas levels 3 and L come closer, as the reverse bias is increased. From these results, we conclude that photogenerated electrons in QW_R are excited to level 3 by the CO_2 laser light.

Electrons in level 3 can relax to level L as well as to level R. The relaxation probability p_{3L} to level L is given by $p_{3L}=\tau_{3R}/(\tau_{3R}+\tau_{3L})$, where τ_{3L} (τ_{3R}) is the scattering time from level 3 to level L(R). These scattering times are of the order of 1 ps and depend on the spatial overlap of the wave function of level 3 with QW_L and QW_R. In the present structure, the wave function of level 3 has a large presence probability in QW_L, and p_{3L} should be close to 1. Electrons that relax to level L recombine radiatively with holes in QW_L, giving rise to ITIL.

According to this transfer mechanism, ITIL intensity is proportional to the concentration of accumulated carriers. The intensity of ITIL peaks depends on bias as shown in Fig. 3. This gives insight into the process of carrier accumulation. Between -1 and -1.5 V, the peak does not shift, but its amplitude increases, indicating the carrier concentration increases, and screens the external electric field. When the bias becomes more negative, the peak shifts and its amplitude saturates, showing the electric field increases, as the carrier concentration saturates for the given Ar^+ laser intensity.

The population of electrons and holes generated by an Ar^+ laser pulse in CTDQWs decays with time. This process was investigated by measuring ITIL intensity as a function of time delay Δt between the turn-off point of Ar^+ laser and the onset point of CO_2 laser pulse. The result is shown in Fig. 4. For $\Delta t>0$, a slow, exponential decrease of ITIL intensity is observed, indicating the carrier concentration decays with a lifetime of 44 μs. This long lifetime insures the efficient and persistent formation of spatially separated carriers and is essential for the appearance of a delayed luminescence. When the CO_2 laser pulse was incident during the Ar^+ laser excitation (-33 μs$<\Delta t<0$), a steep increase of ITIL was observed, illustrating the buildup of separated carrier concentration.

ITIL is a novel and promising mechanism for the conversion of MIR light into NIR or visible light. Delayed luminescence may be used as a novel way to detect MIR by interband photodetectors. To achieve a high conversion effi-

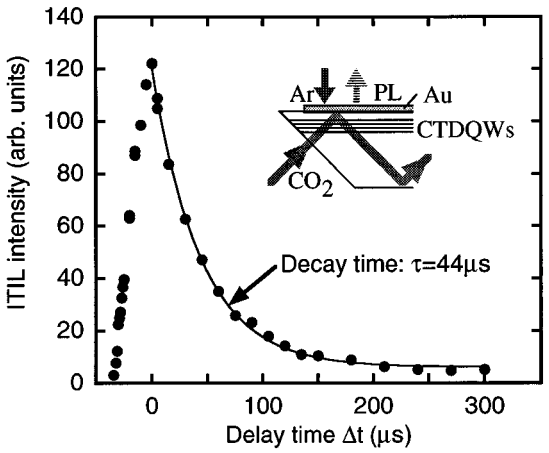

FIG. 4. The time integrated intensity of ITIL induced by a CO_2 laser pulse applied some time Δt after the end of the Ar^+ laser pulse. Experimental conditions are the same as Fig. 2. The circles denote data points and the solid line is an exponential fit for $\Delta t > 0$, with a decay time of 44 μs. The inset shows the sample configuration.

ciency of MIR photon to NIR photon, both the total absorbance of MIR light and the relaxation probability p_{3L} between levels 3 and L must be maximized. Since the absorbance can be made arbitrarily large by increasing the total carrier concentration and/or the length of the waveguide, the maximum conversion efficiency is limited by p_{3L}. In controlling p_{3L} by the structure design of the sample, one must take into account a trade-off relation between p_{3L} and the oscillator strength for intersubband transition between levels R and 3. In efficiently designed structures, a conversion efficiency close to 50% can be achieved when $p_{3L}=0.5$.

In conclusion, we have demonstrated that band-to-band luminescence can be efficiently controlled by a MIR-induced intersubband process in novel double quantum well structures. We proposed to use this phenomenon for MIR to NIR photon conversion. ITIL measurements were also shown to be a sensitive way of probing carrier concentration.

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