Occupation Modulation of Higher Subbands in a Three-Barrier Tunnelling Structure with a Magnetic Field *

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We verify that the magnetic suppression of intersubband LO or LA phonon scattering can give rise to a noticeable nonthermal occupation in higher-lying subbands. This is clearly determined by the relative intensity ratio of the interband photoluminescence spectra for the $E_2 - HH_1$ and $E_1 - HH_1$ transitions. The observed phenomenon may provide an effective method to control the intersubband scattering rate, which is a key factor of the so-called quantum cascade lasers. This is helpful for the population inversion between both the subbands in quantum wells.

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The electrical injection of carriers into higher-lying subbands by sequential resonant tunnelling has proven to be a very effective way for directly observing intersubband spontaneous emission from semiconductor superlattices,[1] nonthermal and inverted population between subbands in a variety of heterostructures (such as superlattices,[2] p–i–n double-barrier structures (DBS),[3,4] coupled quantum wells (QWs)[5] and triple-barrier tunnelling structures[6] (TBTS)), eventually giving birth to a new class of unipolar semiconductor lasers that are the so-called quantum cascade (QC) lasers.[7] It has been a well-established fact that the degree of population inversion between subbands, i.e. the optical gain of subband emission, is very sensitively dependent on the relevant tunnelling rates, intersubband scattering rates in heterostructures, the electrical injection level, and the design of layer structures. However, the possible influence of a perpendicular magnetic field (in the growth direction) on the population inversion between the subbands has not yet been examined, while a depopulation effect by an in-plane magnetic field on the higher-lying subbands in QW structures has been well recorded in the literature.[8−10] This is manifested by observed negative magnetic resistance[8,9] or by the suppression of photoluminescence intensity of the transition from an excited electronic subband to fundamental heavy-hole level.[10]

In this Letter, we verify that a steady-state nonthermal occupation of higher-lying subbands can be created as a result of repopulation of injected electrons between ground and excited subbands in the incident QW of a special TBTS by applying a perpendicular magnetic field. When the TBTS is biased at the peak-current position of the fundamental resonance, its photoluminescence (PL) spectra have several distinct features. With increasing magnetic field, the PL peak exhibits a tremendous redshift (maximally 36 meV) and eventually evolves into three well-resolved PL peaks. Among them, the peak on the lowest energy side traces the redshift of the $E_1 - HH_1$ transition with the magnetic field, which is mainly attributed to the depopulation of the ground state E_1. A newly-appearing PL peak in the middle is believed to come from the $E_2 - HH_1$ transition, indicating a steady-state nonthermal population in the excited subband E_2. We discover that the suppression of LO and LA phonon-mediated intersubband scattering by a perpendicular B-field (about 1.5 T) can be very helpful for establishing a nonthermal or even inverted population between subbands in QWs.

In the experiment, we use a specially designed TBTS grown by molecular beam epitaxy (MBE). A 1.2 μm thick GaAs buffer that is Si-doped to 1.6×10^{15} cm^{-3} is grown on a (100) oriented n$^+$-GaAs substrate, and then followed by a DBS composed of a 2.5 nm thick AlAs barrier, a 7.5 nm thick GaAs well, and a 5.0 nm thick AlAs barrier in sequence. On top of these is a 25 nm thick GaAs well (incident well), covered by a triangle-like $Al_xGa_{1-x}As$ barrier with a mole fraction x graded from 0.8 to 0.1 over a thickness of 20 nm. The top contact layers consist of a 300 nm thick n$^+$-$Al_{0.1}Ga_{0.9}As$ layer, Si-doped to 1×10^{18} cm^{-3}, and a 100 nm thick GaAs with Si doping of 4×10^{18} cm^{-3}.

The sample is processed into a circular mesa of 2 mm in diameter with an annular metal contact formed by evaporating and subsequently alloying Au/Ge/Ni metals. In the magneto-PL experiment,

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the sample is placed in the middle of a superconductor magnet that is immersed in liquid helium. The photoexcitation, which is provided by 488 nm of an Ar^+ ion laser and is kept at the power level of $500\,mW/cm^2$, is guided to the surface of the sample by a fibre. Through the same fibre, the PL signal from the sample is led out to the spectrometer.

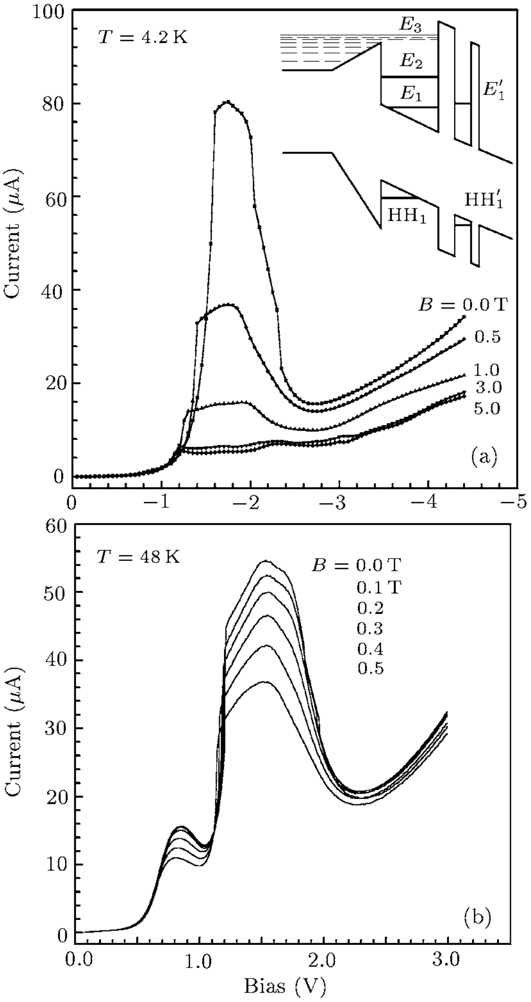

Fig. 1. Drastic suppression of tunnelling current by perpendicular magnetic fields. (a) $I-V$ curves measured in the different magnetic fields up to 5.0 T at a temperature of 4.2 K. The inset shows the band edge profile of the device under negative bias. (b) $I-V$ curves measured in the different magnetic fields up to 5000 G at an elevated temperature of 48 K.

In our previous work,[11] we have measured the $I-V$ characteristics at 4.2 K under different magnetic fields. We have shown that the resonant tunnelling current between the ground subband (E_1) in the wide incident QW and that in the narrow central QW (E_1') is rapidly depressed by increasing B-field only up to 3.0 T, as seen in Fig. 1(a). The similar depression is also observed in the resonant tunnelling between the E_2 and E_1' subbands by applying magnetic fields, as shown in Fig. 1(b). Here the $I-V$ characteristics are measured in the different magnetic fields up to 5000 G at an elevated temperature of 48 K. The weaker current peak appears on the lower bias side to the main peak, stemming from the resonant tunnelling between E_2 and E_1'.

To confirm the assignment, we make a double check for the subband splitting, ΔE_{12}, between E_2 and E_1. This may be estimated in the following two manners. In order to bring the E_1 and E_1' subbands into resonance, a total bias of 1.7 V is consumed to compensate for the energy difference, 48.8 meV, between E_1' and E_1 (E_1 = 7.4 meV, E_2 = 29.5 meV and E_1' = 56.2 meV, obtained under a flat band condition). From the voltage interval of 0.9 V between the first and second current peaks (see Fig. 1(b)), a simple estimation of ΔE_{12} can be given by $\Delta E_{12} = 48.8\,meV \times 0.9\,V/1.7\,V = 25.8\,meV$. On the other hand, a calculation based on the effective mass approximation also shows a value of 27.2 meV for ΔE_{12} under the resonance condition for E_1 and E_1'. One can find a close agreement between these calculated results, which convinces us of the above assignment for the first current peak.

The mentioned novel feature of $I-V$ curves under magnetic fields in the figures has already given a clear clue to the depopulation in the E_1 and E_2 subbands by applying a perpendicular magnetic field. In addition, it is noticed that the E_2 subband may easily be populated at a temperature as low as 20 K and its population is rather sensitive to the magnetic field.

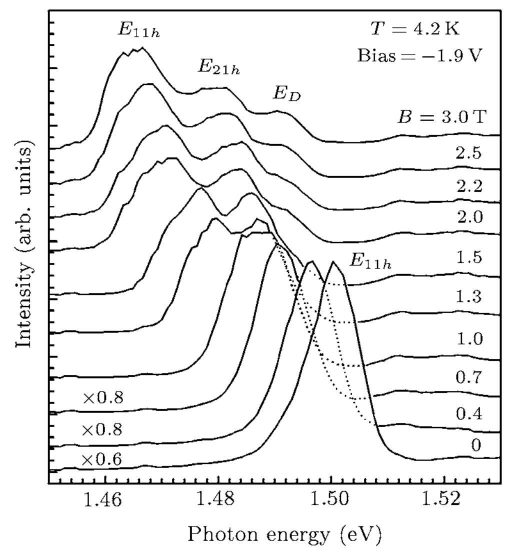

Fig. 2. PL spectra at different magnetic fields for the $E_1 - HH_1$, $E_1 - HH_1$ and $C(D^0 - A^0)$ transitions.

At present, we concentrate on the results of interband PL experiments. The low-temperature (4.2 K) PL spectra obtained as a function of B-field at the bias of −1.9 V (the peak-current position of the fundamental resonance) are plotted in Fig. 2. The main PL peak, appearing at 1.50 eV in zero B-field, can be easily assigned as the recombination of E_1 electrons with HH_1 holes. A slight redshift from the expected energy position 1.524 eV could come from the possible build-in potential due to the large asymmetry in the N–i–N structure itself. Compared with that at zero bias, the E_{11h} peak has already been broadened owing to the filling of the E_1 subband by injected electrons. With increasing B-field, the E_{11h} peak starts to redshift, and gradually evolves into three well-resolved PL peaks. To trace properly the variation of PL peaks with B-field, we adopt a phenomenological fitting procedure of the PL line shapes. As a result, the energy positions of the three PL peaks are plotted as a function of B-field in Fig. 3.

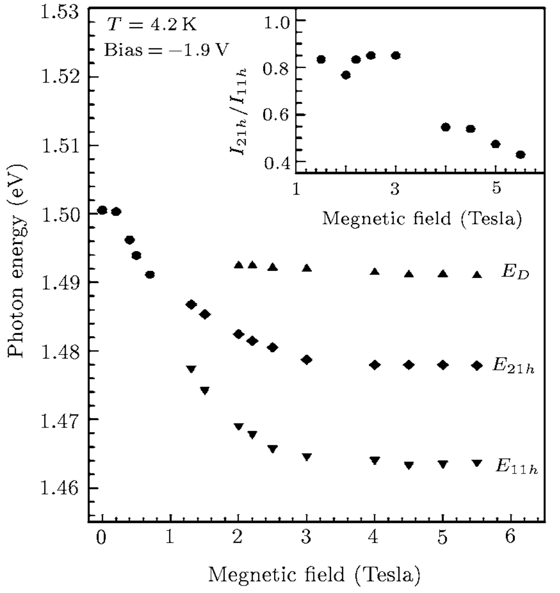

Fig. 3. Dependences of E_{11h}, E_{21h} and E_D peak positions on the magnetic field. The inset shows the relative ratio of integral intensity for E_{11h} and E_{21h} PL peaks.

The peak on the higher energy side, which is absent in zero field, appears as the B-field goes beyond 2 T, and reveals only a slight redshift from 1.492 eV to 1.491 eV. By noticing a 1.2 μm thick GaAs buffer layer of 1.6×10^{15} cm^{-3} Si-doping enclosed in our sample layer structure, one can assign this PL peak to the $C(D^0 - A^0)$ carbon donor–acceptor transition (E_D). An expected increase in the binding energy of the impurities in the presence of the B-field stems naturally from the shrinking of the electronic wavefunction at the impurity site leading to the observed weak redshift. This also explains why its intensity is strengthened in the B-field.

The peak on the lower energy side traces the redshift of the $E_1 - HH_1$ transition in the incident QW with the B-field. Except for a reduction in the PL intensity as a consequence of depopulation of the E_1 subband, an extraordinary redshift of about 36 meV maximally also features our PL data in the presence of the B-field. Because there is a 1.2 μm thick weakly doped GaAs buffer in our sample, an increased resistance of the central TBTS in accordance with the observed suppression of the resonant tunnelling current in the B-field should enhance its voltage sharing in the total voltage drop across the sample. Moreover, the larger the voltage share of the central TBTS, the less conductive the central TBTS becomes due to the further departure from the fundamental tunnelling resonance condition. This gives rise to a positive feedback-like mechanism which may cause a significant potential redistribution in the structure with varying B-field and in turn lead to a quantum confined Stark effect (QCSE) related to the tremendous redshift.

As seen clearly from Figs. 2 and 3, the most intriguing feature of the PL spectra show that a new peak between two former peaks shows up when the B-field goes beyond 1.3 T, and follows the redshift of the $E_1 - HH_1$ transition in the B-field in a smaller magnitude. It is separate from E_{11h} in high B-fields by about 14 meV, which is smaller than the value of $\Delta E_{12} = 25$ meV evaluated previously in the absence of magnetic field and photoexcitation. Such a discrepancy can partially be attributed to the space-charge effect caused by light. In our previous paper,[12] it was verified that the subband separation ΔE_{12} can be tuned to increase substantially by applying a perpendicular electrical field in the same structure. However, the screening of the electrical field in the incident well by photoexcited electrons and holes has a trend of reducing ΔE_{12}. Also, the complex influence of the magnetic field may account for the left discrepancy; for example, the repopulation between the subbands, caused by the variation in intersubband relaxation rates.

From the above facts, we believe that the new PL peak (E_{21h}) arises from the recombination of the electrons in the E_2 level with the holes in the HH_1 level. The PL intensity ratio I_{21h}/I_{11h} between E_{21h} and E_{11h} transitions is plotted as a function of B-field in the inset of Fig. 3. Below 1.5 T one is not able to extract reliable values of I_{21h}/I_{11h} because the two PL peaks are not well resolved. Beyond 3 T intensity ratio I_{21h}/I_{11h} starts to die out. It can be determined that no further change in the occupation of E_1 level should occur when the B-field becomes larger than 3 T, as evident from the measured $I - V$ curve under higher B-fields. However, the PL intensity is also affected by the possible variation of the oscillator strength with the B-field. For lower bounds, the ratio of E_2 and E_1 populations, n_2/n_1, is roughly estimated to be of the order of magnitude of 0.2 from the PL intensity ratio, after correcting for the relative oscillator strengths of the E_{21h} and E_{11h} transitions at the bias of −1.9 V. Differing from many previous works,[3−6] such a steady-state nonthermal population between the subbands in a QW is apparently established by applying a perpendicular B-field.

To give a physical insight into the problem, one should first be aware of the peculiar nature of the transport process in our TBTS. As verified previously,[11] the vertical transport in the structure can be decomposed into two sequential processes. The injected electrons from the heavily doped $Al_{0.1}Ga_{0.9}As$ layer pass over the lowered triangle barrier, and temporarily fill in higher-lying subbands in the incident QW, denoted by E_3 in the inset to Fig. 1(a). The majority of these electrons rapidly cascade down to the first excited subband E_2 and the ground subband E_1 by intersubband relaxation. They eventually escape out of the incident QW by tunnelling through the DBS. As elucidated by theoretical calculation,[11] a perpendicular B-field can make the intersubband relaxation slow down, which in turn affects significantly the relative population in the E_3, E_2 and E_1 subbands. This fact is first registered by the drastic suppression of the resonant tunnelling current peaks with increasing B-field only up to about 2 T (see Fig. 1(a)). However, the possible feedback mechanism mentioned previously may obscure the population changes in the ground and higher-lying states. The PL spectra measured here in the presence of the B-field when the TBTS structure is biased under the resonant tunnelling condition give more direct evidence for the nonthermal occupation in the excited subband E_2, induced by a perpendicular B-field.

The main issue here is the magnetic field dependence of intersubband scattering time $\tau_{21}(B)$, which can be evaluated theoretically by employing the well-established procedure as described, for example, in Refs. [13] and [14]. The calculated results of $\tau_{21}(B)$ for both the electron–LO phonon and electron–LA phonon scatterings show a tremendous magnetic suppression of the intersubband scattering rate by more than four orders of magnitude (Fig. 3),[11] providing a sound grounds for the physical mechanism involved. Obviously, the control of the intersubband scattering rate by applying the B-field will play an important role in establishing the population inversion between subbands in QWs.

In conclusion, the magnetic suppression of LO and LA phonon-mediated intersubband relaxation has been used, for the first time, to establish steady-state nonthermal (even very possibly inverted) population between subbands in QWs, as proven by the interband PL spectra under magnetic fields when the TBTS is biased under the resonant tunnelling condition. Our work may provide an effective method for controlling the intersubband scattering rate, a central issue in quantum cascade lasers, and facilitating the population inversion between subbands in QWs.

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