PHYSICAL REVIEW B VOLUME 56, NUMBER 12 15 SEPTEMBER 1997-II

Magneto-oscillations in the electroluminescence intensity of resonantly coupled GaAs-AlAs superlattices: The magnetoexciton resonance

D. Bertram* and K. v. Klitzing
Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, D-70569 Stuttgart, Germany

O. Kuhn,[†] D. K. Maude, and J. C. Portal
CNRS-LCMI, Avenue des Martyrs 25, F-38030 Grenoble, Cedex 9, France

H. T. Grahn and K. H. Ploog
Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5-7, D-10117 Berlin, Germany

(Received 27 May 1997)

The dependence of the electroluminescence (EL) signal from excited-state interband transitions on the applied magnetic field has been investigated for two superlattice samples. Both samples show clear oscillatory behavior of the EL intensity as a function of the perpendicular magnetic field. In the sample with a subband splitting of the lowest conduction band states above the longitudinal-optical (LO) phonon energy in GaAs, the oscillations are due to the magnetophonon effect. However, in the sample with a subband splitting below the LO-phonon energy of GaAs, two series of oscillations exist, which are assigned to a different relaxation mechanism, the magnetoexciton resonance. The intersubband scattering process is enhanced at certain magnetic-field strengths due to the scattering of free carriers by excitons. [S0163-1829(97)50836-8]

Resonant tunneling in semiconductor heterostructures has attracted much interest in recent years, since it is based on a purely quantum-mechanical effect. At the same time, devices based on resonant tunneling have been proposed, such as the resonant tunneling diode. Furthermore, these systems also have a potential for optical applications.[1,2] It has been recently demonstrated that resonant tunneling in a specially designed superlattice (SL) structure can be used as an optical emitter in the midinfrared region.[3] An important feature of all these devices is the different intersubband relaxation processes which can occur when carriers are electrically injected into a higher subband. In particular, the relaxation channels in the absence of longitudinal-optical (LO) phonon emission are not fully understood and remain to be further clarified.

In this paper, we present evidence for an additional channel of energy relaxation—the magnetoexciton resonance. It appears in the interband electroluminescence intensity of higher subbands, when free carriers and excitons are present at the same time. It becomes the dominating relaxation channel, when relaxation by optical phonon emission is not possible, i.e., for a subband spacing below the optical phonon energy. In samples with a larger subband spacing than the optical phonon energy, the well-known magnetophonon effect is observed.

We have investigated two GaAs/AlAs SL's with different energy spacing of the first two conduction subbands. In sample 1 (2) the width of the GaAs well was chosen to be 13.7 nm (22 nm) so that the subband spacing was above (below) the optical phonon energy $\hbar\omega_{\mathrm{LO}}$ in GaAs (36 meV). Both SL's were grown by molecular-beam epitaxy on n^+ (001)GaAs using 40 periods and an AlAs barrier width of 2.2 nm. The SL's were embedded in the intrinsic region of a p-i-n diode with p and n contact layers of $Al_{0.5}Ga_{0.5}As$ in order to be transparent to the optical emission of the SL interband transitions. After growth the diodes were mesa-etched using wet chemical etching and electrically contacted by evaporating Cr/Au contacts with a diameter of 70 μm on top of the mesas, leaving a large fraction of the mesa surface uncovered, and AuGe/Ni contacts on the substrate side. The bonded structures were mounted in a continuous helium flow cryostat, which was inserted into the 50 mm bore of a resistive, Bitter-type magnet with a maximum field of 23 T. The sample temperature was adjusted to about 5 K. The magnetic field was applied perpendicular to the layer planes of the SL, i.e., parallel to the direction of the SL periodicity. The applied voltage was tuned to the first conduction subband resonance $C2\rightarrow C1$ in the forward bias direction in order to electrically inject electrons and holes into the SL region. The electroluminescence (EL) signal from the SL was dispersed by a 0.64 m monochromator and recorded using an optical multichannel analyzer.

By applying an electric field to the SL, the selection rules for interband transitions are modified. Additional recombination channels, which are forbidden at $F=0$, become allowed at finite electric fields. Detailed investigations of the EL emission as a function of applied voltage at zero magnetic field have already revealed the interband transitions, which become allowed and detectable.[4] The resonance voltages, which are determined by the subband spacing of confined states in adjacent wells, have also been measured.[4,5] At these resonance voltages the transport of electrons or holes is enhanced. In order to investigate the intersubband relaxation properties of these systems, the applied voltage was adjusted to the resonance field $F_{21}=(E_{C2}-E_{C1})/(ed)$, where E_{Cl} denotes the energy of the lth conduction subband and d the SL period. The band-structure profile of part of the SL is schematically shown in Fig. 1 for F_{21}. The investigated interband transition $C2H1$ is indicated. The EL intensity of $C2H1$ was recorded as a function of the magnetic-field strength. The resonance voltage of $C1\rightarrow C2$ did not depend on the applied magnetic field.

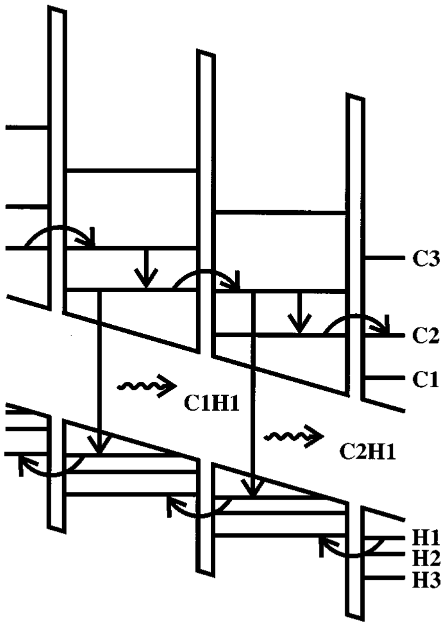

FIG. 1. Schematic diagram of the band-edge profile in the SL under forward bias without any magnetic field. The applied voltage corresponds to the resonance field strength for alignment of the first and second conduction subband between adjacent wells. The investigated interband transition is indicated.

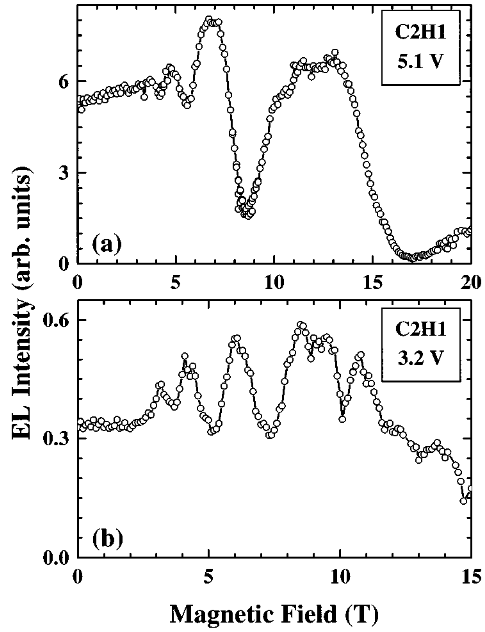

FIG. 2. Electroluminescence intensity of the $C2H1$ interband transition in sample 1 (a) and sample 2 (b) as a function of the applied magnetic field at 5 K. The applied voltage was adjusted to the corresponding $C2\rightarrow C1$ resonance value (5.1 V in sample 1 and 3.2 V in sample 2).

The intensity of the $C2H1$ interband transition is determined by the interplay between tunneling and relaxation. The first process supplies carriers into the confined states, whereas the second process empties the levels under investigation. By fixing the applied voltage to the $C1\rightarrow C2$ resonance value, a fast transport of carriers into the higher conduction band states is ensured. Applying a magnetic field perpendicular to the layer plane of the quantum wells, Landau levels emerge from the quasibound states with a magnetic-field B-dependent energy of $E_i(B)=(i+\frac{1}{2})\hbar\omega_c$, where $\omega_c=eB/m^*$, with m^* denoting the effective mass of the respective carrier in the host material and i an integer 0,1,2 . . . . Thus, the total energy of the different confined states of the SL is a sum of the confinement energy of the lth state without magnetic field E_l and the Landau energy $E_i(B)$. With increasing magnetic field, higher Landau levels of the first subband cross the lowest Landau level of the second subband, resulting in resonances, which are periodic in $1/B$.[6] The corresponding resonance condition reads as follows:

$$\Delta E_{ClC1}=i\frac{\hbar eB}{m^*}, \quad i=1,2,\ldots, \quad l=2,3,\ldots, \tag{1}$$

neglecting effects of nonparabolicity on the effective mass. These direct resonances have already been investigated and are well understood.[7,8] However, additional resonances appear if the total energy of a lower confined state matches the sum of the lowest Landau-level energy of a higher subband plus the LO-phonon energy. This process is referred to as the magnetophonon effect and has been reviewed in Ref. 9. The resonance condition for this resonance is given by

$$\Delta E_{ClC1}-\hbar\omega_{\mathrm{LO}}=i\hbar\omega_c. \tag{2}$$

In sample 1 the subband spacing $\Delta E_{C2C1}=69.6$ meV exceeds $\hbar\omega_{\mathrm{LO}}$ in GaAs so that carrier relaxation via LO-phonon emission is always possible. This process is known to be very efficient.[10] The EL intensity of the $C2H1$ transition at the resonance voltage of 5.1 V in sample 1 is shown as a function of B in Fig. 2(a). Well-defined oscillations with an increasing amplitude are visible. In sample 2 the subband spacing $\Delta E_{C2C1}=31.2$ meV is smaller than $\hbar\omega_{\mathrm{LO}}=36$ meV so that the intersubband relaxation process can be investigated in the absence of LO-phonon emission. The intensity of the $C2H1$ interband transition at the resonance voltage of 3.2 V is shown as a function of magnetic-field strength in Fig. 2(b). The EL spectra have been published previously in Ref. 11. Again, a clear oscillatory behavior is observed. However, the EL transition was not detectable above 15 T. Furthermore, the oscillations in sample 2 do not appear to be periodic in $1/B$.

In order to resolve the different behavior in the two samples, we plot the inverse magnetic-field values of the minima of the EL intensity as a function of the Landau index i. As shown in Fig. 3(a), the data points for sample 1 can be fitted with a straight line intersecting the origin. Applying Eq. (2) using the reduced effective exciton mass with a value of $0.0583m_0$ in GaAs, the energy difference of the states involved in the relaxation process is determined to be $\Delta E=33.6$ meV, which is much smaller than the intersubband spacing $E_{C2}-E_{C1}$ in sample 1. Taking the difference between $E_{C2}-E_{C1}$ and the experimentally determined value, we obtain 36.0 meV, which agrees very well with $\hbar\omega_{\mathrm{LO}}$ in GaAs. Thus, electrons injected into the lowest Landau level of the $C2$ state exhibit an enhancement in the relaxation process by LO-phonon emission, when the difference in Landau levels corresponds to the subband spacing reduced by

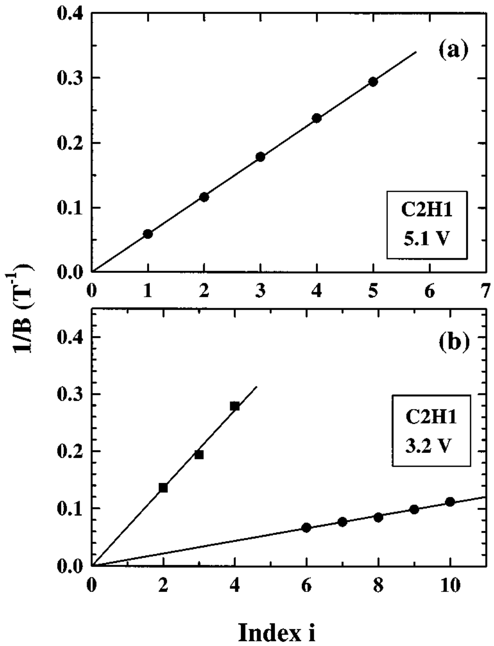

FIG. 3. Inverse magnetic-field values of the minima of the EL-intensity oscillations in Fig. 2 as a function of Landau index i in sample 1 (a) and sample 2 (b). The solid lines are linear fits to the data points.

the optical phonon energy. As a result, the occupation of $C2$ by electrons is decreased, and the recombination of electrons and holes as measured by the EL signal from the $C2H1$ transition is reduced. This effect has also been recently observed in a double-barrier resonant tunneling structure.[12]

For sample 2, relaxation by optical phonon emission is not possible. As shown in Fig. 3(b), two series of oscillations exist in sample 2 in contrast to sample 1. The two branches can be fitted by straight lines intersecting the origin. The first series gives an energy splitting of 25.4 meV using the effective electron mass $m_e^* = 0.067m_0$ in GaAs, while the second series results in an energy spacing of 23.4 meV using the heavy-hole effective mass $m_h^* = 0.45m_0$ in GaAs. Both energy values agree within the experimental error. The subband spacing $E_{C2} - E_{C1}$ in sample 2 has a value of 31.2 meV at the $C1C2$ resonance field strength. Taking again the difference between $E_{C2} - E_{C1}$ and the experimentally determined value, an inelastic scattering process with an average energy of approximately 7 meV has to be taken into account. Since the energy separation of all involved subbands is below the LO-phonon energy of GaAs, no LO-phonon assisted scattering processes can occur in sample 2.

The main scattering mechanisms which can influence the intersubband relaxation in sample 2 are either acoustic-phonon or carrier-carrier scattering. However, the interaction of electrons with acoustic phonons is very small.[13] Only heavy holes couple effectively to acoustic phonons via the deformation potential. Thus, carrier-carrier scattering remains as the only explanation of the oscillatory structure in the EL emission of the excited-state transition in sample 2. However, the characteristic energy cannot be explained by scattering mechanisms between identical carriers such as electron-electron or hole-hole scattering. It must be combined with a bound state of well-defined energy. In large magnetic fields, bound electron-holes states, i.e., excitons, are formed. Therefore, excitons as well as free electrons and holes coexist in the SL region. The existence of excitonic recombination is shown by evaluating the emission energy as a function of magnetic-field strength. An overall quadratic shift is observed with a coefficient similar to the one that has been published to be typical for excitonic emission.[14] Thus, a scattering process between free carriers tunneling into the higher subband and excitons could provide the 10 meV energy loss for the relaxation process.

It is important to note that this resonance appears only in magneto-optical experiments, where the formation of excitons is possible. It has been observed before in transport experiments under photoexcitation, but without clearly identifying the underlying process.[15,16] The value of about 7 meV seems to be reasonable for the excitation energy of an exciton subjected to an electric or magnetic field, as has been shown theoretically by Bauer and Ando.[17] They calculated the excitation energy from the $1s$ to the $2p^-$ state to be weakly magnetic-field dependent and of the order of 7 meV. However, a detailed theoretical description of the behavior of excitons in simultaneous electric and magnetic fields is not yet available. A somewhat similar mechanism was found some time ago in bulk semiconductors. In the so-called magnetoimpurity resonance (MIR), a free electron scatters off an electron bound to an impurity. The binding energy of the electron bound to the respective impurity is used in this scattering process. In our case, a free carrier scatters off an exciton. Therefore, we call this effect the magnetoexciton resonance (MER).

In summary, we have investigated the transport and relaxation processes in GaAs-AlAs superlattices for different subband spacings by means of magnetoelectroluminescence measurements of excited-state interband transitions. For a subband spacing above the optical phonon energy, carriers can effectively scatter using LO phonons. However, for subband spacings below the optical phonon energy, a new relaxation channel is observed. It has been identified as a carrier-carrier scattering process, where free carriers and excitons are simultaneously present in the structure. In analogy to the well-known MIR, this new resonance phenomenon observed in the EL emission of the excited-state transition has been named MER.

We would like to thank A. Fischer for sample growth and L. Eaves for stimulating discussions. This work was supported in part by the European Community ESPRIT Project No. 7193 PARTNERS.

*Present address: NREL, 1617 Cole Boulevard, Golden, CO 80401.

†Present address: NBI, Univ. København, Universitetsparken 5, DK-2100 København Ø, Denmark, and Dept. of Physics, CUT and Univ. Göteborg, S-412 96 Göteborg, Sweden.

[1] C. van Hoof, J. Genoe, R. P. Mertens, E. Goovaerts, and G. Borghs, Electron. Lett. **28**, 123 (1992).

[2] Z. H. Yan, E. Goovaerts, C. van Hoof, and G. Borghs, Superlat-

tices Microstruct. **16**, 239 (1994).
[3] J. Faist, F. Capasso, D. L. Sivco, C. Sirtoni, A. L. Hutchinson, and A. Y. Cho, Science **264**, 533 (1994).
[4] R. Klann, H. T. Grahn, and K. Ploog, Phys. Rev. B **50**, 11 037 (1994).
[5] D. Bertram, H. Lage, H. T. Grahn, and K. Ploog, Appl. Phys. Lett. **64**, 1012 (1994).
[6] J. H. Smet, Ph.D. thesis, Massachusetts Institute of Technology, 1995.
[7] L. Eaves, G. A. Toombs, F. W. Sheard, C. A. Payling, M. L. Leadbeater, E. S. Alves, T. J. Foster, P. E. Simmonds, M. Hennini, O. H. Hughes, J. C. Portal, G. Hill, and M. A. Pate, Appl. Phys. Lett. **52**, 212 (1988).
[8] M. L. Leadbeater and L. Eaves, Phys. Scr. **T35**, 215 (1991).
[9] L. Eaves and J. C. Portal, J. Phys. C **12**, 2809 (1979).
[10] K. T. Tsen and H. Morkoç, Phys. Rev. B **34**, 4412 (1986).
[11] D. Bertram, H. T. Grahn, O. Kuhn, D. K. Maude, J. C. Portal, K. Ploog, and K. von Klitzing, Surf. Sci. **361/362**, 387 (1996).
[12] P. D. Buckle, J. W. Cockburn, M. S. Skolnick, R. Grey, G. Hill, and M. A. Pate, Phys. Rev. B **53**, 13 651 (1996).
[13] M. Cardona (private communication).
[14] L. Ma, K-S. Lee, C. H. Perry, J. M. Worlock, and J. E. Golub, in *Proceedings of the 20th International Conference on the Physics of Semiconductors*, edited by E. M. Anastassakis and J. D. Joannopoulus (World Scientific, Singapore, 1990), p. 1297.
[15] M. C. Smith, A. Petrou, C. H. Perry, and J. M. Worlock, Surf. Sci. **174**, 136 (1986).
[16] W. Müller, H. T. Grahn, K. v. Klitzing, and K. Ploog, Phys. Rev. B **48**, 11 176 (1993).
[17] G. E. W. Bauer and T. Ando, Phys. Rev. B **38**, 6015 (1988).