Enhancement of the photoluminescence intensity in short-period GaAs/AlAs superlattices with different well and barrier thickness

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The transition from an indirect to a direct energy band structure has been induced in short-period GaAs/AlAs superlattices by going from a symmetric to an asymmetric distribution of the well and the barrier thickness within the unit cell of the superlattice. Reducing the barrier thickness d_B to half the well thickness d_W moves the lowest state in the conduction band from the X point in the AlAs barrier to the Γ point in the GaAs well. For $d_W = 2d_B$, the band structure becomes therefore direct for all values of d_W. This change in the type of energy gap is accompanied by a significant enhancement of the integrated photoluminescence intensity for asymmetric superlattices. © *1999 American Institute of Physics.* [S0003-6951(99)03817-6]

Short-period GaAs/AlAs superlattices (SLs) have been shown to exhibit an indirect band structure for well thicknesses below 3.4 nm (12 monolayers), if well and barrier thickness are equal.[1–5] This transition is caused by the staggered alignment (type II) of the SLs band structure, when the X minimum of AlAs in the conduction band is lower than the respective Γ state in GaAs. In this case, the electron-hole transitions become indirect in both, real and momentum space. Since indirect-gap semiconductors have inferior light emitting properties than their direct-gap counterparts, short-period GaAs/AlAs SLs have not been used for light-emitting devices, although their energy gap falls in the red part of the visible spectral region.

A closer look at the energy band properties of short-period GaAs/AlAs superlattices reveals that there is a mixing between the X states of AlAs and Γ states of GaAs in the conduction band due to potential fluctuations at the interfaces. For well thicknesses greater than 0.85 nm, this mixing results in a quasi-direct energy gap, i.e., the absorption or emission of electrons at the energy gap may occur without momentum-conserving phonon participation.[4,5] Nevertheless, weak phonon replicas below the dominant luminescence line are still observed in the emission spectra of quasi-direct SLs. However, for indirect SLs (i.e., a well thickness $\leqslant 0.85$ nm), the emission spectra are completely dominated by phonon-assisted transitions.[6,7] The luminescence properties of direct-gap SLs are therefore superior for two reasons. First, the oscillator strength, which is determined by the overlap of the electron and hole wave function, is much larger for direct than for quasi-direct or indirect transitions. Second, the direct-gap emission spectra do not contain any phonon replicas, i.e., they consist of a single line. Both properties result in a substantial increase of the luminescence efficiency in structures with a direct energy gap compared to indirect or quasi-direct energy-gap short-period SLs.

For SLs with a well thickness below 3.4 nm, a reduction of the barrier width for a fixed well width can eventually result in a change of the band alignment from quasi-direct to direct.[8] However, this effect was only studied for a particular well width so that no general conclusions for short-period SLs with a well width below 3.4 nm could be drawn. Furthermore, the effect of the change of the type of band alignment on the emission intensity was not investigated in detail.

In this letter, we will show that, by choosing the ratio of well and barrier thickness to be larger than unity, short-period GaAs/AlAs SLs with arbitrary well width can exhibit a direct energy-gap band structure. If the thickness ratio is two or larger, short-period GaAs/AlAs SLs with any well thickness below 3.4 nm will always exhibit a direct energy gap, resulting in a significant enhancement of the photoluminescence (PL) intensity. SLs with an equal well and barrier thickness are referred to as symmetric SLs, while all others are asymmetric SLs.

We have investigated the low-temperature PL properties of a number of short-period $(GaAs)_n/(AlAs)_m$ SLs, where n and m denote the well and barrier widths, respectively, in monolayers (ML). The samples labeled as n/m are divided into two groups, symmetric SLs with $n = m$ and asymmetric SLs with $n \neq m$. The sample parameters are listed in Table I.

TABLE I. Well thickness n in ML, ratio n/m of well to barrier thickness m, number of periods N, ground state energy of Γ state in well E_0^Γ and X state in barrier E_0^X, energy difference $\Delta E = E_0^\Gamma - E_0^X$, and type of energy gap for the investigated samples.

Sample	n	n/m	N	E_0^Γ (meV)	E_0^X (meV)	ΔE (meV)	type
10/10	10	1	100	221	201	20	II
7/7	7	1	150	314	221	93	II
5/5	5	1	150	386	246	140	II
7/5	7	1.4	50	290	247	57	II
6/4	6	1.5	50	308	269	39	II
10/5	10	2	50	200	247	−47	I
8/4	8	2	50	236	270	−34	I
6/3	6	2	200	271	302	−31	I

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All samples were grown by molecular beam epitaxy on semi-insulating (100) GaAs substrates. In most cases, the SL period and the composition have been confirmed by x-ray diffractometry. For the 6/3 sample, the difference between the nominal and actual layer thicknesses was about 1%. For the other samples, the disagreement between nominal and actual layer thickness was usually less than 10%.

The PL experiments were performed at 4.2 K using a cw Ar^+ laser operating in all-lines mode for excitation. The PL spectra were recorded using a 0.5 m monochromator and a photomultiplier with an overall spectral resolution of less than 1 meV.

The calculated energies for the lowest electron state in the GaAs well and AlAs barrier are also listed in Table I. The calculations have been performed using the well-known Kronig–Penney model. The used effective masses for electrons in GaAs and AlAs at the Γ point in units of the free electron mass m_0 were 0.0665 and $0.15m_0$, respectively, and at the X point 1.3 and $1.1m_0$, respectively. The value of the conduction band offset between AlAs and GaAs at the Γ point (X point) was 0.982 eV (-0.175 eV). The widths of the layers for the symmetric SLs are all smaller than 12 ML, which corresponds to the thickness of the transition from a direct-gap to an indirect-gap band structure so that the symmetric SLs exhibit an indirect energy gap. This implies that the lowest state in the conduction band is at the X point of the AlAs barrier, i.e., the band alignment is type II. Going from the symmetric to the asymmetric SLs, the well thickness was kept approximately constant, while the barrier width was reduced. This approach was chosen in order to remain for the symmetric SLs in the regime of indirect energy gaps. By reducing the barrier width by a factor of 2 with respect to the well width, the lowest state in the conduction band occurs now for all well thicknesses at the Γ point of the GaAs wells, i.e., they become type I. However, for a thickness ratio smaller than two, the asymmetric SLs can still be indirect.

The normalized PL spectra of the symmetric SLs are shown in the lower part of Fig. 1, while the corresponding spectra of the asymmetric SLs with $n=2m$ are displayed in the upper part. The PL spectra of the symmetric SLs exhibit an intensive zero-phonon line caused by the recombination of spatially indirect excitons consisting of X_z electrons in the barrier and heavy holes at the Γ point in the well. In addition, there are weak phonon replicas on the low-energy side of the PL line connected with GaAs- and AlAs-like interface phonons.[7,9] These spectra are typical for quasidirect gap, symmetric SLs with $3<n<12$. For $n \leqslant 3$, the symmetric SLs become fully indirect with the conduction band minimum at the $X_{x,y}$ point of AlAs.[6,7]

Enlarging the PL signal of the 10/10 SL on the high-energy side reveals an additional line separated by 56 meV from the zero-phonon line. It is probably due to the direct Γ–Γ transitions in the well region. The calculated X_z–Γ splitting of 20 meV listed in Table I is somewhat lower, but this discrepancy could be removed by assuming a barrier thickness 1 ML smaller than the nominal one.

The PL spectra of the asymmetric SLs with $n=2m$ consist of a single line, which for the 10/5 and 8/4 are blueshifted with respect to the position of the corresponding symmetric SLs (cf. inset of Fig. 1). These PL spectra are characteristic for direct-gap SLs, for which the excited electrons and holes thermalize in the same layer (GaAs) and recombination occurs through Γ levels in conduction and heavy-hole valence bands. Increasing the amplification of the PL signal on the low-energy side does not reveal any additional lines, which can be assigned to phonon satellites. At the same time, the energy distance of 16 meV between the PL maximum of the asymmetric SL 10/5 and Γ line of the symmetric SL 10/10 agrees well with the calculated shift of 21 meV (cf. Table I) for the Γ–Γ transitions with a decreasing barrier width from ten to five monolayers. Note that due to the stronger coupling in the 10/5 SL, which implies a larger miniband width, the direct energy gap of the 10/5 SL lies below the one of the 10/10 SL. Due to the very large miniband width in the 6/3 SL, the direct energy gap of this sample is actually close to the one of the corresponding symmetric SL. Therefore, the indirect energy gap of the 5/5 SL is at a higher energy than the direct gap of the 6/3 SL. We conclude that the investigated asymmetric SLs with $n=2m$ exhibit a direct band structure, i.e., they are of type I. The calculations of the ground state energies show that all asymmetric SLs with $n=2m$ have a direct energy gap, since with decreasing barrier width the lower edge of the miniband at the Γ point in the well increases less strongly than the blueshift of the ground state at the X point in the AlAs barrier.

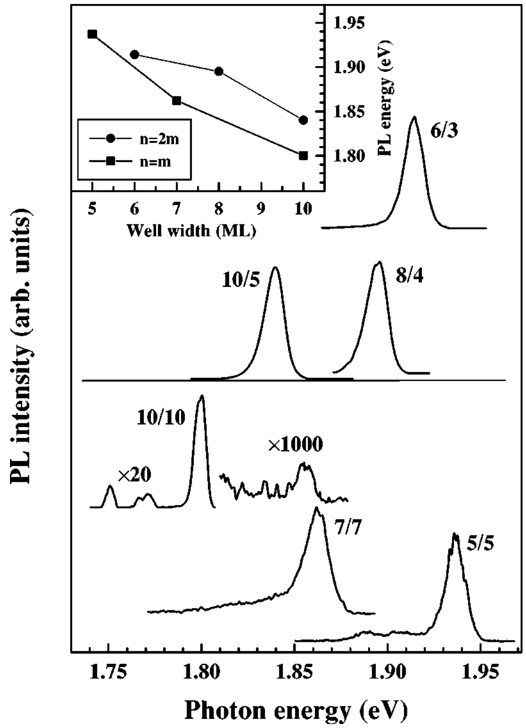

FIG. 1. PL spectra of the symmetrical (lower part) and asymmetrical (upper part) SLs recorded at 4.2 K. The spectra of the different samples were normalized to their respective maximum intensity. The inset shows the energetic positions of the maximum PL vs the well width.

We investigated some additional asymmetric SLs with a thickness ratio of about 1.5 as listed in Table I. The PL spectra of the 7/5 and 6/4 asymmetric SLs contain the typical zero-phonon line and phonon replicas of SLs with a quasidirect band structure. The calculations confirm this assignment. Even a 5/3 SL would still be indirect or very close to

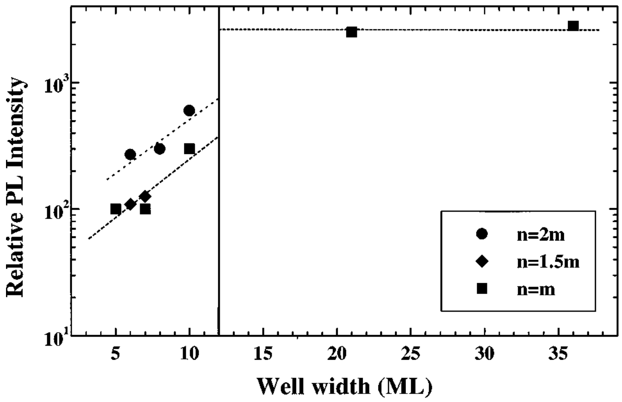

FIG. 2. Integrated PL intensity vs well width for the symmetric (squares), asymmetric with $n=1.5m$ (diamonds), and asymmetric with $n=2m$ SLs (dots). The vertical solid line indicates the well width for the $\Gamma-X$ crossover in symmetric SLs.

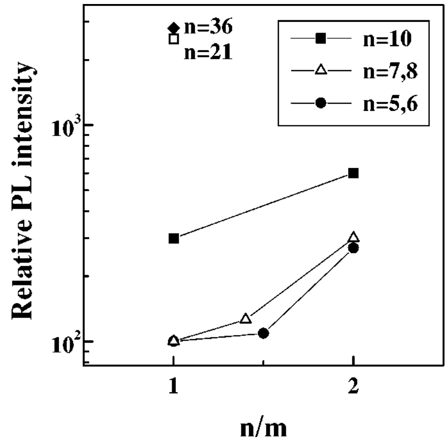

FIG. 3. Integrated PL intensity vs ratio of well-to-barrier width in ML for all samples shown in Fig. 2 as indicated.

the crossover so that in order to ensure the direct nature of the energy gap for asymmetric SLs it is necessary to choose at least a value of 2 for the n/m ratio.

The transition from symmetric to asymmetric SLs for $n<12$ ML is accompanied by a significant enhancement of the PL intensity. In Fig. 2, the integrated PL intensity is shown as a function of the well width for the three symmetric SLs shown in Fig. 1 as well as the asymmetric SLs together with two symmetric SLs with a well width of 20 and 36 ML. The vertical line marks the thickness of the $\Gamma-X$ crossover for symmetric SLs. The integrated PL intensity appears to increase with increasing well thickness. At the same time, the asymmetric SLs with $n=2m$ exhibit an enhancement of the PL intensity by a factor of about 3 over the symmetric SLs and the asymmetric ones with $n=1.5m$, which is caused by the greater probability of spatially direct transitions. This observation may be useful for practical application of the short-period asymmetric SLs in light diodes.

The observed increase of the PL intensity is significantly smaller than one would expect for direct transitions, whose oscillator strength is known to be several orders of magnitude larger than that of the indirect or quasi-direct transitions. Apparently, this reduction of the enhancement is probably due to a stronger influence of the interface corrugations with decreasing well thickness, which results in a great enhancement of nonradiative losses.

In Fig. 3, the integrated PL intensity is plotted versus the ratio n/m. With decreasing well thickness, there appears to be a saturation of the achievable PL signal for both symmetric SLs and asymmetric SLs. We believe that this saturation is due to the increasing influence of interface recombination for thinner well thicknesses. Nevertheless, by using an asymmetric SL with at least $n=2m$ instead of a symmetric one, the intensity can be increased for the same well thickness. Although the direct-gap symmetric SLs have a significantly larger PL intensity than the direct-gap asymmetric ones, changing the growth parameters for the asymmetric SLs may improve the PL efficiency.

In conclusion, short-period GaAs/AlAs SLs with an asymmetric choice of well and barrier thickness can exhibit a direct energy gap in the parameter range, where symmetric short-period GaAs/AlAs SLs are indirect. In particular, when the thickness of the well is at least two times larger than the barrier thickness, asymmetric SLs become direct for all well thicknesses. These asymmetric SLs exhibit superior PL emission properties over their symmetric counterparts. This observation may open new possibilities for using short-period GaAs/AlAs SLs in light-emitting devices in the long-wavelength region of the visible spectrum.

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