Enhancement of electron-phonon interaction in ultrashort-period GaAs/AlAs superlattices

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(Received 4 March 1996)

The interaction of electrons and phonons is strongly enhanced in ultrashort-period GaAs/AlAs superlattices. The enhancement is caused by an increase of the exciton binding energy with decreasing superlattice period. The photoluminescence spectra of indirect-gap superlattices exhibit a zero-phonon line due to scattering by defects and/or interfacial roughness as well as pronounced phonon sidebands. [S0163-1829(97)07015-X]

I. INTRODUCTION

The interaction of electrons and phonons influences the transport as well as optical properties of semiconductors.[1] The conductivity, radiative and nonradiative recombination, spontaneous and stimulated luminescence, and the carrier plasma are strongly affected by electron-phonon scattering. In previous investigations it was shown that in bulk semiconductors the surface can strongly influence the recombination properties and electron-phonon interaction.[2] Under these conditions, photoexcited carriers are mainly scattererd by surface phonons.

In semiconductor quantum wells and superlattices the density of interface states in greatly increased compared to the respective bulk material. In particular, scattering effects in short-period superlattices (SL's) should be dominated by interface states. In previous investigations of the optical properties of short-period GaAs/AlAs SL's, several phonon replicas were observed in photoluminescence (PL) experiments for layer thicknesses below 10 ML.[3–13] However, no detailed investigation of the intensity of the phonon replicas on the layer thickness has been performed, to our knowledge.

In this paper, we demonstrate that the electron-phonon interaction is strongly enhanced in ultrashort-period $(GaAs)_n/(AlAs)_n$ SL's with n denoting the layer thickness in ML. We have investigated the low-temperature PL for superlattices with $n=1,2,\dots,10$. For $n\leqslant 5$, three PL sidebands below the excitonic PL line are observed, which are identified as phonon-assisted recombination lines. These sidebands can be assigned to a zone-folded LA GaAs mode and optical interface phonons of GaAs and AlAs. For $n\leqslant 3$, the phonon-assisted PL lines clearly dominate the spectra. For $n=3$ the energy gap changes from quasidirect to indirect, i.e., the conduction-band minimum occurs at X_{xy} in the AlAs barriers. The observation of the zero-phonon PL line becomes possible due to a breakdown of the momentum conservation rule by interface microcorrugations and/or defects. A broad PL line below the phonon replicas in SL's with $n\leqslant 3$ originates from defects as determined from temperature-dependent PL measurements.

II. EXPERIMENT

The SL samples were grown by molecular-beam epitaxy. The parameters of the different samples are summarized in Table I. In all samples the well and barrier widths were identically given in monolayers of GaAs and AlAs, respectively. In the following the samples will be denoted by n/n. In GaAs and AlAs a monolayer corresponds to a thickness of about 0.283 nm. The samples were excited by an Ar^+-ion laser in all-lines mode. The PL measurements were per-

TABLE I. Well and barrier widths in monolayers, number of periods N, calculated energy gap E_{calc}, measured energy of exciton line E_{expt}, and energy of phonon replicas E_{p1}, E_{p2}, and E_{p3} measured from the exciton line for the investigated GaAs/AlAs superlattices.

Sample	N	E_{cacl} (eV)	E_{expt} (eV)	E_{p1} (meV)	E_{p2} (meV)	E_{p3} (meV)
1/1	800	2.099	2.0615	12.5	27.3	45.0
2/2	400	2.072	2.0570	12.0	29.5	45.0
3/3	250	2.024	2.0463	11.5	28.3	45.8
4/4	200	1.973	2.0260	9.6	30.4	48.4
5/5	150	1.924	1.9407	12.1	28.6	47.8
7/7	150	1.850	1.8760		28.0	48.0
10/10	100	1.789	1.7980		28.0	48.0

formed at 5 K in a He-flow cryostat using a 1-m monochromator (600 lines/mm grating) and a N_2-cooled charge coupled device array for detection. The PL lines were fitted with four Gaussians to obtain their respective position, intensity, and full width at half maximum (FWHM). The calculated energy gap was obtained within a Kronig-Penney model. The effective masses used in GaAs (AlAs) at the Γ point in units of the free-electron mass m_0 were $0.0665m_0$ $(0.15m_0)$ for electrons and $0.377m_0$ $(0.48m_0)$ for heavy holes. At the X point we used an electron effective mass of $1.3m_0$ $(1.1m_0)$. The value of the conduction-band offset between AlAs and GaAs at the Γ point (X point) was 0.982 eV (-0.175 eV). For the valence-band offset we used a value of 0.529 eV.

III. RESULTS

In Fig. 1(a) the photoluminescence spectra recorded at 5 K of five SL's with $n/n = 1/1,\ldots,5/5$ are shown. Note that the intensity of the spectra for the $n/n = 4/4,\ldots,1/1$ SL's have been rescaled by the indicated factor. The overall intensity of the PL lines decreases with decreasing n/n. However, the distribution of the intensity between the different lines strongly changes when n is decreased. Figure 1(b) shows the spectrum of the 3/3 SL including a fit to four Gaussian lines. The identification of the four lines in the figure will be explained below. For all displayed spectra the PL signal can be described by four Gaussian lines having different energies, intensities, and widths. The energy positions obtained from these fits are listed in Table I.

In Fig. 2(a) the position of the line highest in energy is plotted versus the layer width together with the calculated energy gap. As expected, the energy gap increases with decreasing layer thickness reaching a value of 2.099 eV, which is still below the corresponding value of 2.109 eV for the alloy $Al_{0.5}Ga_{0.5}As$. The experimental data display a clear tendency of saturation below $n/n = 4/4$. The validity of a simple Kronig-Penney model has to be questioned in this regime. A more advanced model calculation of the energy gap in this range is necessary to understand the underlying physical process. Nevertheless, this PL line is identified as the indirect excitonic recombination between the X_1 state in the AlAs layer and the heavy-hole ground state HH_1 in the GaAs layer.

In Fig. 2(b) the energy positions of the three lines at lower energies are shown versus the layer thickness. For the samples 7/7 and 10/10 only two, very weak satellites are resolved. The energetic positions of these three lines exhibit a rather weak dependence on the layer thickness. The replica with an energy of 11.5 meV is assigned to a zone-folded LA GaAs phonon based on the observation of these modes in Raman scattering by Spitzer *et al.*[14] in similar samples. Furthermore, zone-folded LA modes have also been observed in magnetotransport measurements of direct-gap GaAs-AlAs superlattices.[15] The other two energies of 28.8 and 46.4 meV are closely related to GaAs and AlAs optical phonons, respectively. However, their energies are considerably lower than those of confined modes originating from bulk LO modes. Therefore, these two energies are assigned to interface phonons, whose energies are typically in this range. For example, an AlAs interface mode with an energy of 46 meV

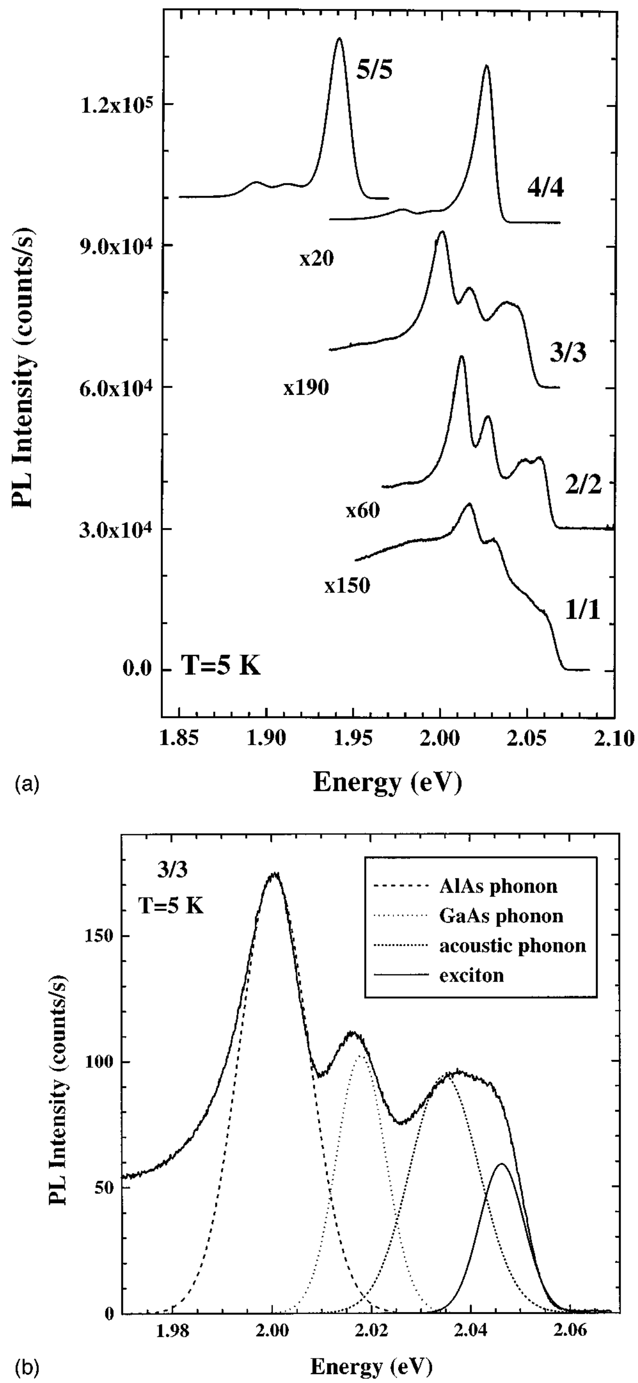

FIG. 1. (a) Photoluminescence spectra of the 1/1, 2/2, 3/3, 4/4, and 5/5 GaAs/AlAs superlattices at 5 K. The spectra for the 1/1, 2/2, 3/3, and 4/4 SL's have been multiplied by 150, 60, 190, and 20, respectively, in comparison with the 5/5 SL. (b) PL spectra of the 3/3 SL including a fit to four Gaussian lines as indicated in the figure.

has recently been observed in Raman scattering experiments.[16] It has also been reported that interface modes couple very strongly to electrons.[17] The increasing density of interface versus bulk states with decreasing n supports this interpretation.

In Fig. 3(a) the relative intensity of the exciton line and its FWHM are plotted versus the layer thickness. The integrated intensity of the PL line is normalized by the total intensity of

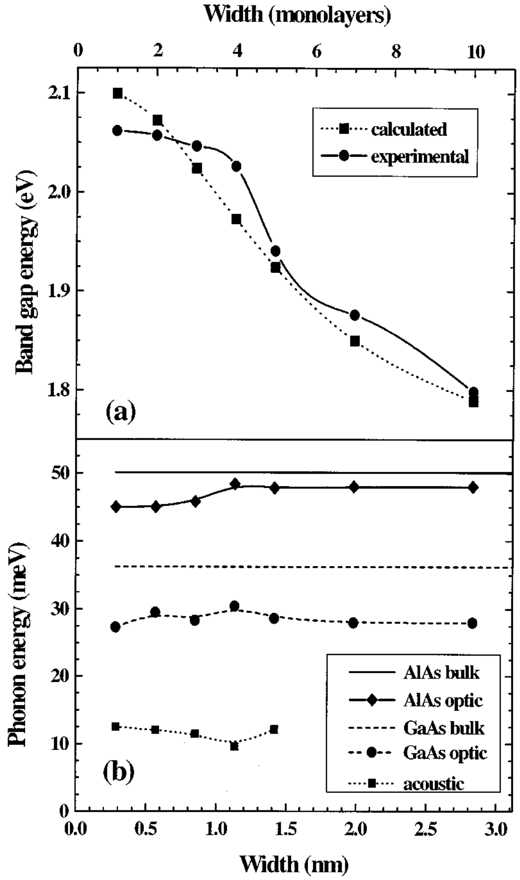

FIG. 2. Energy of the calculated energy gap as well as of the experimentally observed exciton recombination line (a) and energy of the phonon replicas (b) vs the width of the GaAs/AlAs superlattice layers at 5 K. The horizontal lines in (b) indicate the maximum phonon energy in bulk GaAs and AlAs.

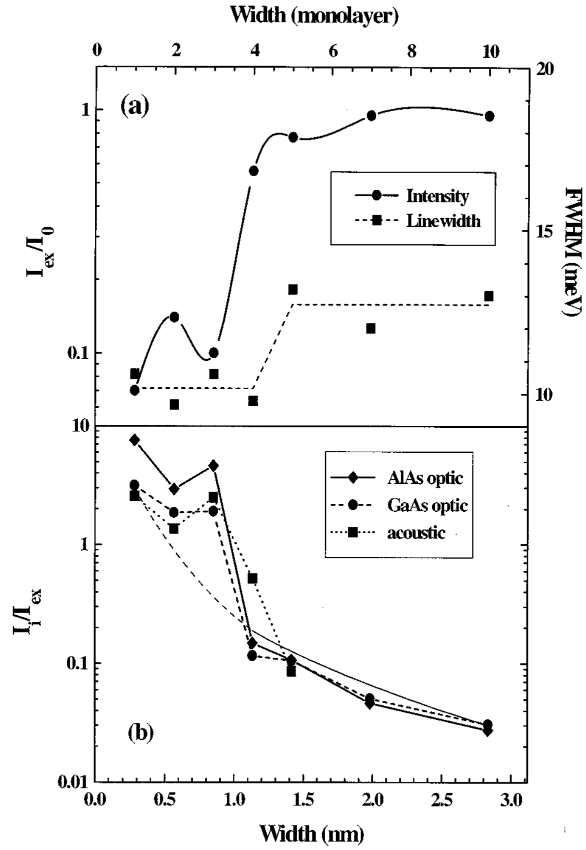

FIG. 3. Intensity normalized to the total intensity I_0 as well as full width at half maximum of the exciton line (a) and strength of interaction of electrons with different phonons I_i/I_{ex} (b) vs the width of the GaAs/AlAs superlattice layers at 5 K. The dashed line in (b) corresponds to a d^{-2} dependence.

all PL lines for the respective samples. There is a clear reduction of the relative intensity of the exciton line by one order of magnitude, when the layer thickness is decreased from $n/n=7/7$ to 3/3. It increases again by a small amount for 2/2, but finally drops to the lowest value for 1/1. At the same time the FWHM is also reduced from a value of about 13 to 10 meV. The relative intensity, which is removed from the excitonic line, is gained by the PL sidebands.

IV. INTERPRETATION AND DISCUSSION

The intensity ratio of the phonon replicas I_i with respect to the exciton line I_{ex}, are a measure for average number of emitted phonons N, which is proportional to the square of the electron-phonon interaction coupling constant $W_{\text{el-ph}}$. If the intensity of the phonon replicas and of the exciton line are normalized to the total intensity I_0, the ratio of the normalized intensities I_i/I_0 and I_{ex}/I_0 is independent of the total intensity and equal to the ratio of the absolute intensities I_i/I_{ex}. The absolute intensities I_i and I_{ex} of different samples depend on too many experimental parameters such as the laser intensity, the number of layers in the SL, the surface layer, etc., so that it is not useful to compare these quantities. We therefore use the direct ratio of these two intensities as a measure for the average number of emitted phonons. It should be mentioned that for quasidirect-gap SL's, i.e., for $4<n<12$, the intensities of the zero-phonon line and the phonon replicas depend on the corresponding matrix elements, which are inversely proportional to the splitting of the of the Γ and X levels.[6] These matrix elements will vary as the period of the SL changes. However, the variation will be the same for the excitonic and phonon-assisted transitions. This conclusion is supported by time-resolved photoluminescence measurements, which show the same lifetimes for the zero-phonon and phonon-assisted lines. In Fig. 3(b) the quantity I_i/I_{ex} is plotted versus layer thickness for different values of i. The interaction constant exhibits a strong increase for all three phonon lines. For $n\geqslant 4$, the dependence of the two optic-phonon lines follows approximately a d^{-2} law.

The increase of $W^2_{\text{el-ph}}$ for $n\geqslant 4$ in the case of the LO-phonon replicas can be connected with the increase of the exciton binding energy with decreasing layer thickness. Using the qualitative expression for the Fröhlich interaction $W^2_{\text{el-ph}}$ as obtained by Hopfield,[18] the coupling can be expressed as

$$W^2_{\text{el-ph}} \propto \frac{1}{a_B^* E_{\mathrm{LO}}}\left(\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon}\right), \tag{1}$$

where a_B^* denotes the effective Bohr radius using the effective masses and dielectric constants of the respective material, E_{LO} the LO-phonon energy, ϵ_∞ the high-frequency dielectric constant, and ϵ the low-frequency dielectric constant. Relating the effective Bohr radius to the exciton binding energy E_{ex} via $a_B^* = e^2/(8\pi\epsilon_0\epsilon E_{ex})$ leads to

$$W_{\text{el-ph}}^2 \propto \frac{E_{ex}}{E_{LO}}\left(\frac{\epsilon}{\epsilon_\infty}-1\right). \quad (2)$$

Assuming the typical dependence of $E_{ex} \propto d^{-2}$, the measure for the electron-phonon interaction decreases as d^{-2}. Our interpretation is supported by calculations of the exciton binding energy in short-period SL's, which show a strong increase with decreasing layer thickness.[19,20] However, this dependence only applies for layer thicknesses corresponding to the pseudodirect energy-gap regime, where the conduction-band minimum is formed by the X_z state of the AlAs layers. For $n \leqslant 3$, GaAs/AlAs SL's exhibit a true indirect energy gap, i.e., the conduction-band minimum occurs for X_{xy} states in the AlAs layers.[21] In this case a large wave-vector zone-boundary phonon is needed to assist the recombination. In indirect-gap SL's, zero-phonon lines are observable because of the scattering of carriers by imperfections in the SL such as defects, microcorrugations, etc. The appearance of the strong increase of the phonon-assisted PL lines over the zero-phonon line may also be due to an enhanced electron-phonon interaction. However, to extract the electron-phonon coupling constant in the case of indirect-gap SL's, it is necessary to develop a full microscopic model of electron states, phonon modes, and interfacial roughness and defects for $n \leqslant 3$.

The spectrum of the 1/1 SL is dominated by a large increase of the PL intensity below the phonon sidebands. This strong increase is probably due to a strong increase in the density of defects. However, since the strong increase of the phonon sidebands occurs between $n=4$ and 3, where this low-energy PL is much weaker, the effect of the relative increase of the phonon sideband PL over the exciton PL cannot be explained considering only a strong increase in defect PL. A detailed modeling of this SL system is necessary to determine, which effect is stronger, the increase in electron-phonon coupling or the increase in defect PL.

V. SUMMARY AND CONCLUSIONS

In summary, the interaction of electrons and phonons in ultrashort-period GaAs-AlAs superlattices strongly increases with decreasing layer thickness. In the quasidirect regime for $n \geqslant 4$, where the conduction-band minimum is formed by an X_z state of the AlAs layer, the coupling constant increases approximately as d^{-2} with decreasing thickness d. This increase is attributed to the enhancement of the exciton binding energy. A very pronounced increase of the relative intensity of the phonon-assisted PL lines in the SL's with $n \leqslant 3$ is due to the transition of the band structure of the SLs from quasidirect to indirect. The observation of the exciton line in this regime could be a result of enhanced exciton scattering by microcorrugations and defects. For $n=1$, the PL spectrum is dominated by a broad, low-energy line which probably originates from an enhanced density of defects.

ACKNOWLEDGMENTS

We would like to thank H.-P. Schönherr for sample growth and M. Nakayama (Osaka City University) for stimulating discussions. This work was supported in part by NATO and the Ukrainian Council of Science and Technology.

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