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Effects of confinement on the electron–phonon interaction in $\text{Al}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs}$ quantum wells

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Abstract

Photoluminescence measurements at different temperatures have been performed to investigate the effects of confinement on the electron–phonon interaction in GaAs/AlGaAs quantum wells (QWs). A series of samples with different well widths in the range from 150 up to 750 Å was analyzed. Using a fitting procedure based on the Pässler- p model to describe the temperature dependence of the exciton recombination energy, we determined a fit parameter which is related to the strength of the electron–phonon interaction. On the basis of the behavior of this fit parameter as a function of the well width thickness of the samples investigated, we verified that effects of confinement on the exciton recombination energy are still present in QWs with well widths as large as 450 Å. Our findings also show that the electron–phonon interaction is three times stronger in GaAs bulk material than in $\text{Al}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs}$ QWs.

1. Introduction

The built-in barriers originated by the superposition of semiconductor materials with different energy gaps lead to a one-dimensional size quantization that confines electrons, holes, and excitons within the semiconductor with the smaller energy gap. The reduction of the system dimensionality from the three-dimensional (3D) case for bulk materials to the quasi-bi-dimensional (Q2D) case for quantum wells alters a number of basic physics properties of the heterolayers and a detailed understanding of the changes is required for the implementation of exciton-based devices, such as thermovoltaic solar cells [1], field-effect transistors [2], and photodetectors [3].

In the present work, we are, in particular, concerned with the investigation of the effects of the confinement on the

electron–phonon interaction in GaAs/AlGaAs quantum wells. The reduction of the dimensionality of a system leads to significant changes on the electron–phonon interactions and, consequently, to modifications of the transport and optical properties of the semiconductor heterolayers [4, 5]. Typically, excitons are confined in QWs when the quantum well width (L_w) is about or smaller than the exciton diameter, $2a_0$, where a_0 is the exciton Bohr radius [6, 7]. For GaAs, using the relative dielectric constant $\epsilon = 12.4$, and the reduced effective mass $\mu^* = 0.056m_0$ (m_0 is the free electron mass) [8], we obtain $a_0 \approx 120$ Å, which means that confinement effects on the exciton recombination energies are not expected for QWs larger than about 240 Å.

For most semiconductor materials and heterostructures, the exciton recombination energy, E_x , decreases with the increase of the temperature due to the electron–phonon

interaction and to the thermal expansion of the lattice. At low temperatures ($T \leq 100$ K) the electron–phonon interaction is mainly determined by longitudinal acoustic (LA) phonons [9–15]. In this same temperature range, in general, the thermal expansion of the lattice is very small and its influence on the decrease of E_X with temperature can be neglected [9, 10, 16]. For instance, for GaAs and $\text{Al}_x\text{Ga}_{1-x}\text{As}$ materials, the thermal dilatation of the lattice leads to a reduction of only $\sim 2\%$ of E_X in the temperature range $0 \text{ K} < T \leq 100 \text{ K}$ [17]. Due to this small contribution, several models describing the temperature dependence of E_X were proposed in the literature taking into account the electron–phonon interaction but neglecting the contribution of the thermal expansion of the lattice [18]. Among them we find the semi-empirical model proposed by Viña *et al* [19], which uses the Bose–Einstein statistical distribution of energy for the phonon modes, and more recent analytical models such as the ones proposed by Pässler [12, 20].

Studies performed on $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ QWs comparing several theoretical models usually used to describe the temperature dependence of the exciton recombination energy indicate that the Pässler- p model is more appropriate for obtaining information about confinement effects in these systems. In general, the fit parameters of the Pässler- p model are strongly dependent on the Al concentration of the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ barrier material [13, 18, 21, 22]. For instance, the parameter related to the mean energy of the phonons in the temperature scale increases its magnitude with the increase of the Al concentration in the range $0 \leq x \leq 0.45$ [21, 23, 24]. Lourenço *et al* [21, 22] also showed that, in the temperature range $12 \text{ K} \leq T \leq 300 \text{ K}$, the introduction of the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ barriers in $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ QWs reduces the values of the parameters of the Pässler model with increase of the Al concentration in the range $x < 0.20$. For $x \geq 0.20$ the parameters have their values increased with increase of the Al concentration as observed for $\text{Al}_x\text{Ga}_{1-x}\text{As}$ bulk material, and for $x \approx 0.40$ the parameters surpass the values obtained for GaAs bulk [21, 22, 25]. Another work investigating the temperature dependence of E_X , in the temperature range $12 \text{ K} \leq T \leq 100 \text{ K}$, in 60 \AA thick $\text{AlGaAs}/\text{GaAs}$ QWs showed that confinement effects lead to an abrupt decrease of the Θ_p and α_p parameters of the Pässler- p model as compared to the GaAs bulk values [18].

Since this theoretical model has parameters that are related to the strength of the electron–phonon interaction, if we measure the temperature dependence of the exciton recombination energy in QWs with different well widths, we will be able to obtain information about the effects of the confinement on the electron–phonon interaction. So, in the present work we measured by means of photoluminescence (PL) spectroscopy the temperature dependence of the exciton recombination energy in $\text{Al}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs}$ QWs for the temperature range $12 \text{ K} \leq T \leq 100 \text{ K}$. Performing a fit procedure based on the temperature dependence of E_X , the parameters related to the Pässler- p model were determined for each sample of the set. In order to analyze the effects of confinement on the electron–phonon interaction, the parameters determined for the QWs and for the GaAs bulk

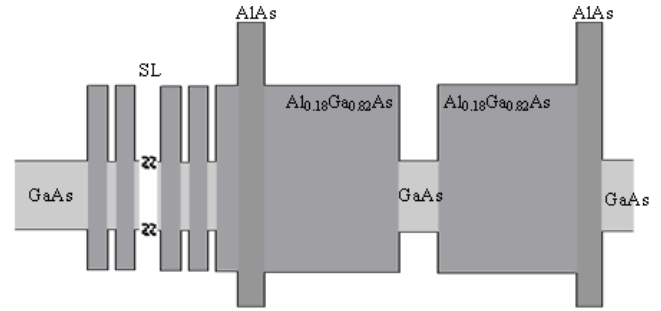


Figure 1. Schematic representation of the conduction and valence band edges of the samples.

material were compared. We verified that confinement effects on the exciton recombination energy are still present in QWs with well widths as large as 450 \AA . Our findings also showed that the electron–phonon interaction is three times stronger in GaAs bulk material than in $\text{Al}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs}$ QWs.

2. Samples and experimental details

All the samples here investigated were grown in a Gen II MBE system on epitaxially semi-insulating GaAs(001) substrates. The structure consisted of a $1 \mu\text{m}$ thick GaAs buffer layer followed by a $30 \times [\text{Al}_{0.18}\text{Ga}_{0.82}\text{As}(100 \text{ \AA})/\text{GaAs}(50 \text{ \AA})]$ superlattice, the quantum well region, and a final 50 \AA GaAs layer. The GaAs quantum well was surrounded by two 500 \AA thick $\text{Al}_{0.18}\text{Ga}_{0.82}\text{As}$ barriers containing two 20 \AA thick AlAs layers symmetrically located at the end of each well barrier. Five samples were grown, each one with a different value of the quantum well width: 150 \AA (#2553), 300 \AA (#2554), 450 \AA (#2555), 600 \AA (#2557), and 750 \AA (#2558). A schematic representation of the conduction and valence band edges of the heterostructure is shown in figure 1. The reference structure (sample #2409) consisted of a $4 \mu\text{m}$ thick intrinsic GaAs layer grown on top of a buffer layer composed of a $10 \times [\text{AlAs}/\text{GaAs}]$ superlattice grown on top of a semi-insulating GaAs(001) substrate.

The exciton recombination energy experimental data were obtained by PL spectroscopy and measured in the temperature range from 12 up to 100 K . The samples were excited with the 5145 \AA line of an argon laser with a maximum excitation power of 7 mW and a spot size of about $260 \mu\text{m}$ on the surface of the sample. The luminescence signal was analyzed using a monochromator, detected using a GaAs photomultiplier, and the electrical signal was amplified and processed using standard lock-in techniques.

3. Theoretical modeling

For the modeling of the experimental data we used the Pässler model, in which an analytical expression is introduced to fit the dependence of the semiconductor energy gap with temperature given by [12]

$$E_X(T) \cong E_X(0) - \frac{\alpha_p \Theta_p}{2} \left[\sqrt[p]{1 + \left(\frac{2T}{\Theta_p}\right)^p} - 1 \right] \quad (1)$$

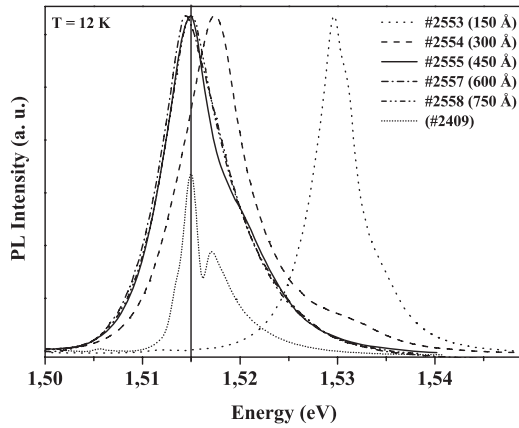


Figure 2. PL spectra of the reference sample (GaAs bulk) and $\text{Al}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs}$ QWs with different well widths, measured at $T = 12$ K. The vertical line indicates the peak position of the heavy-hole–exciton emission of the reference sample.

where $E_X(0)$ is the exciton recombination energy at $T = 0$ K, α_p is the slope of the curve $E_X(T)$ for $T \rightarrow \infty$, i.e., $\alpha_p \equiv -(dE_X(T)/dT)_{T \rightarrow \infty}$. Θ_p is an empirical parameter that can be associated with the effective mean energy of the phonons in the temperature scale. The relation between Θ_p and the mean temperature of the phonons, $\Theta = \langle \varepsilon \rangle / k_B$, is given by the expression

$$\Theta \cong \Theta_p [1.152 + 0.145 \ln(p - 1.7)]. \quad (2)$$

The exponent p controls the spectral function $f(\omega) \propto (\hbar\omega)^\nu$, where $p = \nu + 1$, and is related to the dispersion coefficient Δ given by $\Delta = (p^2 - 1)^{-1/2}$. The relation between the parameters α_p and Θ_p leads to the parameter $a_p = (\alpha_p \Theta_p) / 2$, which is proportional to the strength of the electron–phonon interaction.

In order to calculate the exciton binding energies and to evaluate the dimensionality of the QWs investigated here, we used the fractional-dimensional formalism [26]. In the α -dimensional (α D) space, the exciton binding energy, E_b , can be determined by solving the time-independent Schrödinger equation in a noninteger-dimensional space:

$$\left[-\frac{\hbar}{2\mu^*} \nabla_\alpha^2 - \frac{1}{\varepsilon_r \varepsilon_0} \frac{e^2}{|\vec{r}|} \right] \psi(\vec{r}) = E \psi(\vec{r}) \quad (3)$$

where μ^* is the exciton effective reduced mass, ε_r is the relative dielectric constant, and the Laplacian operator is given by [27]

$$\nabla_\alpha^2 = \frac{\partial}{\partial r^2} + \frac{\alpha - 1}{r} \frac{\partial}{\partial r} + \frac{1}{r^2 (\sin \phi)^{\alpha-2}} \frac{\partial}{\partial \phi} (\sin \phi)^{\alpha-2} \frac{\partial}{\partial \phi} \quad (4)$$

where α is the system dimensionality. The energy eigenvalues of equation (3) in the noninteger space are given by

$$E_b(n) = \frac{4E_{b0}}{(2n + \alpha - 3)^2} \quad (5)$$

where $n = 1, 2, 3$ is the principal quantum number, E_{b0} is the exciton binding energy in the 3D bulk material, and α is

the fractional dimensionality of the effective space that would model the real system. In order to determine the values of α we adopted the analytical expression proposed by Mathieu *et al* [28, 29]:

$$\alpha = 3 - \exp\left(-\frac{L_w^*}{2a_0^*}\right) \quad (6)$$

where a_0^* is the exciton effective Bohr radius and L_w^* the effective well width of the QW. The GaAs low temperature material parameters [8, 30] used in the calculations are as follows: $m_e^*/m_0 = 0.0665$, $m_{hh}^*/m_0 = 0.51$, and $\varepsilon_r = 12.4$. From the results obtained with the fractional-dimensional formalism we determined the exciton binding energies and the dimensionality of the QWs, where α is in the range $2 < \alpha < 3$ for the Q2D systems and $\alpha = 3$ for 3D systems.

4. Results and discussion

Figure 2 shows the PL spectra measured at 12 K for all the samples analyzed in this work. The PL spectra of the samples #2554–#2558 show a main peak assigned to the heavy-hole (hh)–free exciton recombination energy, E_X , associated with the first subband. For the 450 Å thick QW (#2555), $E_X = 1515.1$ meV. For the samples with $L_w \geq 600$ Å, the hh–exciton energy is practically equal to the free exciton emission energy measured for the reference sample (GaAs bulk) at 1514.9 meV. The PL spectra of the 150 Å thick QW (#2553) shows the hh–exciton energy at 1530.8 and at 1529.7 meV a peak related probably to a biexciton emission. We did not observe in the whole set of PL spectra (as a function of temperature or excitation power) any emission associated with the second electronic subband. As a consequence, anomalies such as the ones reported by Pozela *et al* [31] for modulation doped heterostructures showing the inversion of the peak intensities associated with electron optical transitions from the first and second subbands to the valence band in the GaAs quantum for well widths in the range $L_w = 225\text{--}300$ Å do not occur in the undoped heterostructures analyzed here.

In figure 2, effects of confinement can be clearly observed when we compare the exciton recombination energies in the PL spectra of the GaAs bulk material and of the 150 Å thick QW. The effects of confinement in the wavefunction and energy of excitons confined in QWs appear when the well width thickness is comparable to the exciton diameter, i.e., when $L_w \approx 2a_0$. For instance, effects of confinement were observed for excitons confined in GaAs QWs with well widths as large as 300 Å [32, 33], 240 Å [34], and 200 Å [6, 7, 35]. Confinement effects on the exciton recombination energies are not expected for $L_w > 2a_0$, although our results show that these effects are still observed in the exciton energies for the samples with well widths in the range $300 \text{ Å} \leq L_w < 450 \text{ Å}$ (see figure 2).

Combining theoretical and experimental data obtained by means of reflectivity and PL spectroscopy, Kusano *et al* [35] obtained interesting results for $\text{Al}_{0.19}\text{Ga}_{0.81}\text{As}/\text{GaAs}$ QWs. They verified by PL measurements that confinements effects are still present in the energy of excitons in QWs as large as 2010 Å. Reflectance measurements of the same authors also showed confinement effects in samples with 900, 2010 and 5200 Å. They also verified that the exciton emission

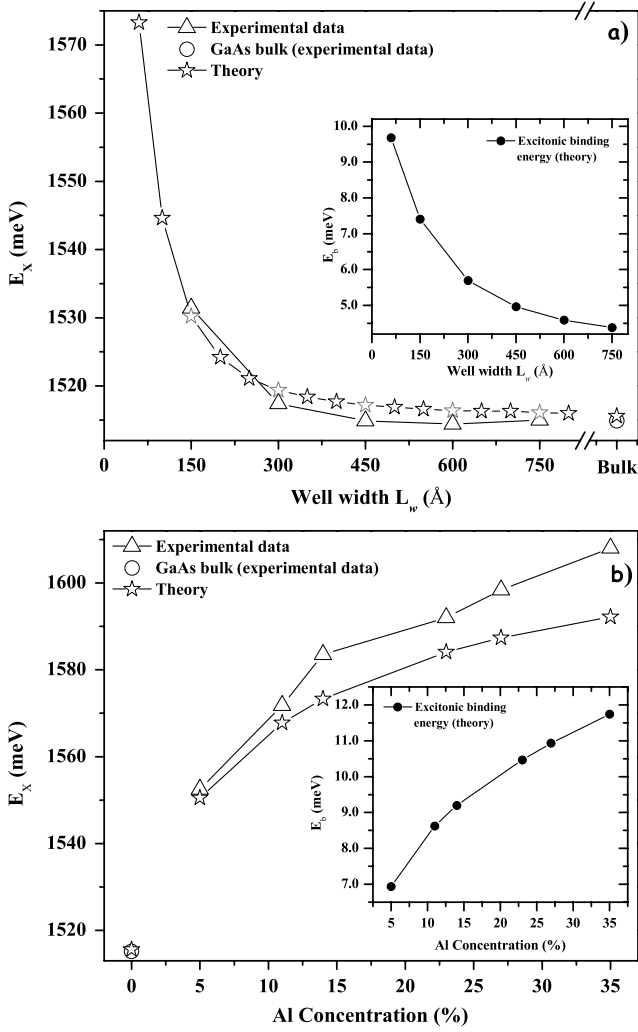


Figure 3. Heavy-hole-free exciton recombination energy: (a) extracted from the PL spectra shown in figure 2, (b) experimental data from [18], and theoretical data of E_X (star symbols) and E_b (sphere symbols) obtained with the fractional-dimensional formalism (see the text).

energies observed in the PL spectra of samples with well widths as large as 990 and 2010 Å under external applied magnetic fields have a behavior which is similar to the exciton emission energy observed in thin quantum wells under equal experimental conditions.

Figure 3(a) shows the experimental values of the exciton recombination energies for different well widths extracted from the PL spectra of figures 2 and 3(b) shows the experimental data published by da Silva *et al* [18] for different values of the Al concentration in 60 Å thick $\text{Al}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ QWs. The theoretical values of E_X in figures 3(a) and (b) were calculated using the expression

$$E_X = E_{n1} + E_{\text{hh1}} + E_g - E_b \quad (7)$$

where E_{n1} and E_{hh1} are the fundamental energy levels of electrons and holes, respectively, confined in a quantum well of finite barrier height [36], $E_g = 1519.2$ meV is the GaAs energy gap at low temperature [8], and E_b is the exciton binding

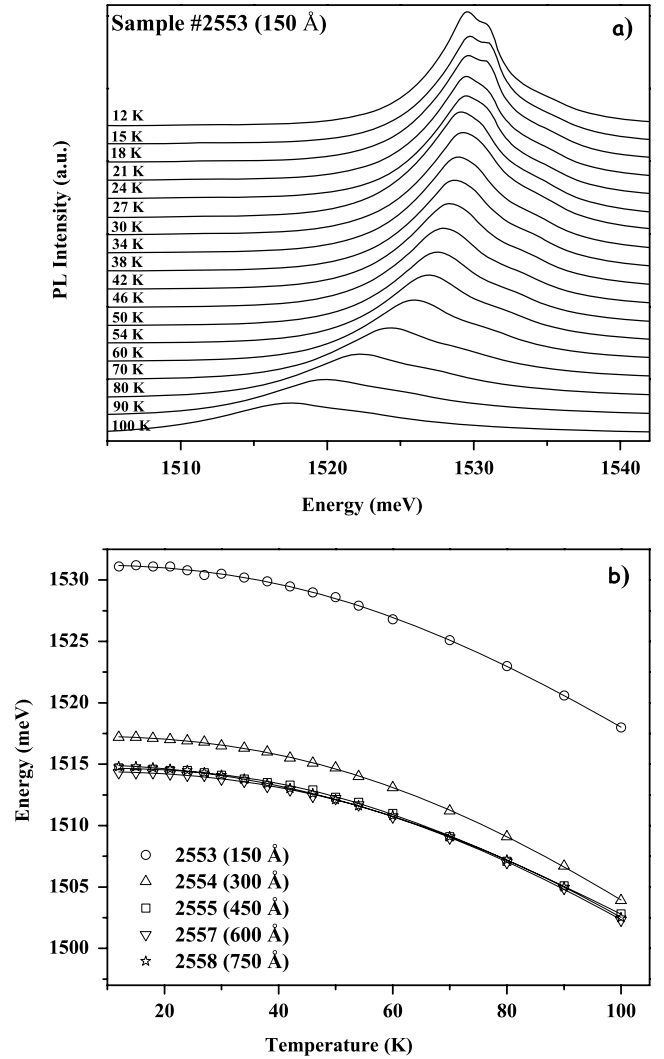


Figure 4. (a) PL spectra of sample #2553 ($L_w = 150$ Å) recorded at different temperatures. (b) Energy chart of the PL peaks related to the heavy-hole-exciton recombination energy as a function of temperature for all the samples analyzed in the present work. The lines connecting the experimental points (symbols) are the best fit curves determined according to the Pässler- p model applied to the $E_X(T)$ experimental data.

energy for the fundamental state ($n = 1$) calculated by means of the fractional-dimensional formalism [28, 29].

Figure 3(a) shows that E_X has a pronounced decrease in the range $150 \text{ Å} \leq L_w \leq 450 \text{ Å}$, decreasing from 1530.8 meV at $L_w = 150 \text{ Å}$ down to 1515.1 meV at $L_w = 450 \text{ Å}$, but has a constant value for the samples with the largest well widths, that is equal to the observed free exciton energy of the reference sample observed at 1514.9 meV. Figure 3(a) also shows that the experimental well width dependence of E_X is well represented by the theoretical results. In figure 3(b), theoretical and experimental results show that E_X increases with increase of the Al concentration. Although the two curves describe the same qualitative trend, the theoretical values are underestimated when compared to the experimental results. The insets in figure 3 show the exciton binding energy values used in equation (7) to calculate the theoretical values of E_X .

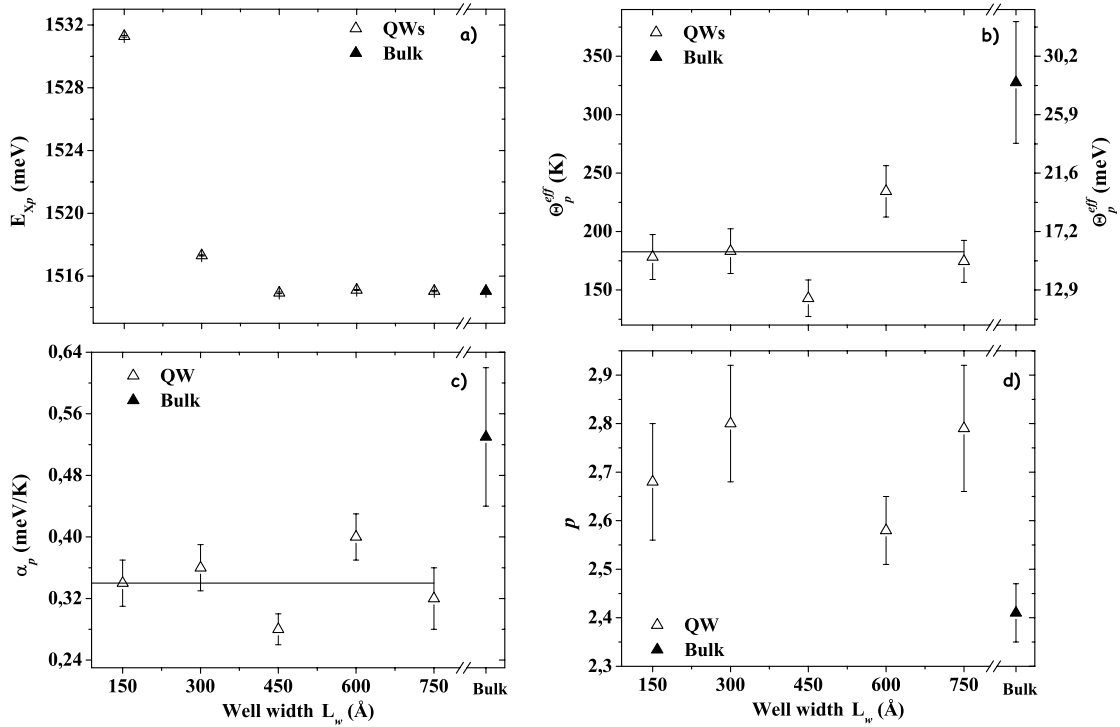


Figure 5. Parameters related to the best fit curves shown in figure 4(b) and for the GaAs bulk material: (a) exciton recombination energy at $T = 0$ K; (b) mean energy of the phonons in the temperature scale; (c) slope of the $E_X(T)$ versus T curve as $T \rightarrow \infty$; (d) p .

Figure 4(a) shows the PL spectra of sample #2553 ($L_w = 150$ Å) recorded at different temperatures. PL measurements as a function of excitation intensity (not shown here) and temperature indicate that the peak observed in the PL spectrum recorded at $T = 12$ K on the low energy side around 1529.7 meV is a biexciton emission. The intensity of this peak increases with increase of the excitation intensity but disappears with increase of temperature around $T = 42$ K. The peak observed at 1530.8 meV at $T = 12$ K related to the heavy-hole–exciton recombination and dominates the PL spectra for temperatures $T > 42$ K. The energy and intensity of the exciton peak decrease with increase of the temperature, being observed at 1516.9 meV at $T = 100$ K. A similar behavior of the exciton peak was also observed in the PL spectra of the other samples of the series.

In figure 4(b) we show the exciton recombination energy (symbols) extracted from the PL spectra (not shown here) recorded in the range $12 \text{ K} \leq T \leq 100 \text{ K}$ for each sample of the series, and also the respective best fit curves (full lines) determined according to the Pässler- p model applied to the $E_X(T)$ experimental data. The pronounced shift to higher energy of the $E_X(T)$ curve related to the sample with $L_w = 300$ Å as compared to the curves of the samples with larger well widths ($L_w \geq 450$ Å) indicates that the effects of confinement on the exciton recombination energy are still weighty for QWs with well width thicknesses larger than the values accepted in the literature [6, 7, 32, 33].

In figure 5 we show the values of the parameters that were determined by our fitting procedure using the Pässler- p model to describe the temperature dependence of the recombination energy of excitons confined in QWs with different well widths.

We see that, initially, $E_g(0)$ decreases with increase of L_w , but has a constant value for $L_w \geq 450$ Å (see figure 5(a)). For the parameter Θ_p (see figure 5(b)) we obtained the values of 140 K (12.1 meV) for the 450 Å thick QW and 238 K (20.5 meV) for the 600 Å thick QW. The mean value of Θ_p for the whole set of QW samples is ~ 182.7 K (~ 15.8 meV), which is much lower than the value of 324.7 K (28 meV) that we found for GaAs bulk. We found $\alpha_p \sim 0.26$ meV K $^{-1}$ for 450 Å thick QW and $\alpha_p \sim 0.38$ meV K $^{-1}$ for the 600 Å thick QW (see figure 5(c)). The mean value of α_p for the series of samples is 0.34 and 0.53 meV K $^{-1}$ for GaAs. In figure 5(d) we see that the minimum value of the parameter p is 2.59 for the 600 Å thick QW and the maximum value is 2.82, obtained for 300 Å thick QW. We also found $p = 2.44$ for GaAs bulk. The values of p for the QWs and for GaAs bulk lead to intermediate values of the dispersion parameter (α) that are in the range $0.3 \leq \alpha_p \leq 0.57$, as observed for most semiconductor materials [37].

In figure 6 we show the parameters α (that describes the dimensionality of the system) and a_p (that is related to the strength of the electron–phonon interaction) as a function of the well width thickness. The values of α are 2.54 for the 150 Å thick QW and 2.95 for the 750 Å thick QW, a value that is very close to 3, representing the 3D systems. The parameter a_p has a minimum value equal to 18 meV (for $L_w = 450$ Å) and a maximum of ~ 45 meV (for $L_w = 600$ Å). The mean value of $a_p = 31.6$ meV while the value for GaAs is $a_p = 86.8$ meV, a result which leads us to conclude that the electron–phonon interaction is three times stronger in GaAs bulk than in the Al $_{0.18}$ Ga $_{0.82}$ As/GaAs QWs. We also notice in figure 6 that even for the largest QWs with $\alpha \approx 3$, the presence of the barriers

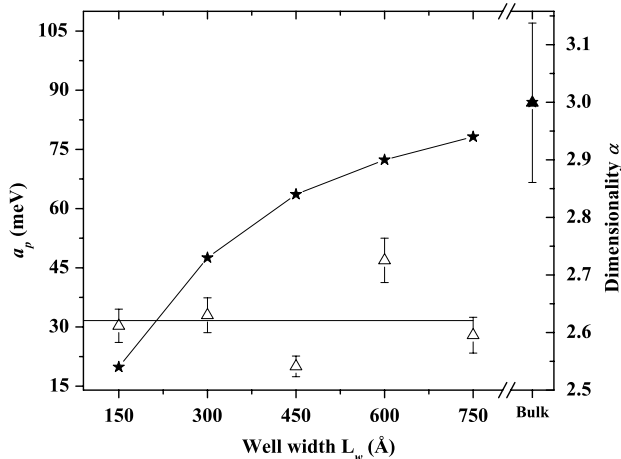


Figure 6. Dimensionality parameter α (star symbols) and the a_p parameter that is related to the strength of the electron–phonon interaction (sphere symbols) as a function of the well width. The lines connecting the points are guides to the eyes.

is not a negligible effect on the electron–phonon interaction. For instance, the largest QW with $\alpha = 2.95$ still has a Q2D characteristic if confinement effects on the electron–phonon interaction are concerned.

Our findings are in agreement with the results reported by Rudin *et al* [38] obtained by the analysis of the temperature dependence of the half-width at half-maximum (HWHM). These authors verified that the electron–acoustic phonon interaction has approximately the same strength in QWs with different well widths ($L_w \leq 200$ Å) but is systematically weaker in GaAs bulk.

For semiconductor bulk materials and heterostructures, the contribution of the exciton–acoustic phonon interaction to the HWHM at low temperature can be described by the expression $\Gamma = \Gamma(0) + aT$, where $\Gamma(0)$ is a term that does not depend on temperature and the parameter a (in units of $\mu\text{eV K}^{-1}$) describes the strength of the electron–acoustic phonon interaction [39]. Experimental data for the HWHM derived from several different spectroscopy techniques applied to the investigation of GaAs QWs with $L_w \leq 200$ Å show that the strengths of the interactions of electrons with acoustic phonons are in the range 1–3 $\mu\text{eV K}^{-1}$ [40–42], while the experimental results for GaAs bulk are in the range 8–12 $\mu\text{eV K}^{-1}$ [38, 43]. Theoretical calculations performed by Piermarocchi *et al* [44] provide values in the range 1.5–2 $\mu\text{eV K}^{-1}$ and the calculations of Rudin *et al* [38] give $a = 2$ $\mu\text{eV K}^{-1}$ for QWs and 7.8 $\mu\text{eV K}^{-1}$ for GaAs bulk material. We see that theoretical and experimental results provide smaller values for the electron–acoustic phonon interaction parameter of AlGaAs/GaAs QWs when compared to the values obtained for GaAs bulk. In addition, our findings also indicate that this reduction of the electron–phonon interaction occurs even for QWs with well width thicknesses as large as 750 Å.

5. Conclusion

We have investigated the effects of confinement on the electron–phonon interaction in a series of $\text{Al}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs}$

QWs with different well widths. We showed that confinement effects are present in QWs with well width thicknesses much larger than the exciton diameter. We also showed that the electron–phonon interaction is three times stronger in GaAs bulk material than in $\text{Al}_{0.18}\text{Ga}_{0.82}\text{As}/\text{GaAs}$ QWs.

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