Temperature and excitation intensity dependencies of the photoluminescence spectra of GaAs/(AlGa)As disordered superlattices

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The temperature and excitation intensity dependencies of the photoluminescence (PL) spectra of GaAs/$Al_{0.3}Ga_{0.7}As$ superlattices (SL's) having randomly distributed well widths are studied. Our results indicate that the electronic properties of disordered SL's are similar to those of other disordered semiconductors: at low energies the electronic states are essentially localized, while at higher energies they are extended. Further, the PL spectra feature a disorder-induced fine structure, and they shift to the red with T simply following the reduction of the band gap. The PL efficiency shows a weaker decrease with increasing T than the ordered SL. The dependence of the PL spectra on the excitation intensity shows an anomalous behavior of the disorder-induced fine-structure recombination lines. [S0163-1829(96)02036-X]

In the last decade, theoretical calculations on random superstructures,[1–3] as well as the development of the techniques for fabricating artificial multilayered materials, strongly stimulated experimental investigations on disordered superlattices (d-SL's). The study of disorder-induced localization effects in semiconductor materials is attracting a renewed interest nowadays.[4–7] Recent experimental results[4] have shown that in short-period d-SL's the decrease of the photoluminescence (PL) efficiency with increasing temperature is slower than in ordered SL's (o-SL's) and bulk alloys. These results stimulated additional efforts devoted to the investigation of electronic and optical properties of d-SL.[5–7]

In the present work we investigate the dependencies on temperature T and excitation intensity I_o of the PL spectra of GaAs/$Al_{0.3}Ga_{0.7}As$ o-SL and d-SL having randomly distributed well widths. SL's ranging from the ordered case to strongly disordered ones are studied. In previous works,[7] we discussed in detail the low-T absorption and reflectance spectra of d-SL's and the predictions of the transfer-matrix method. We report experimental results on the dependence of the PL spectra on T from 2 to 300 K, and on the dependence on I_o over six decades.

We introduce disorder into o-SL's by replacing randomly chosen wells by thicker ones (while the barrier width remains constant), such that for the different well widths present in the now disordered SL we can write

$$L_{Wi}=L_{W\min}+(i-1)\Delta L_W, \quad i\in\{1,..,N_W\}. \tag{1}$$

Thus, there are N_W different widths that differ by the increment ΔL_W. As ΔL_W is increased, we change the degree of disorder according to the Anderson definition.[3,7] We choose a $\Delta L_W \gg 1$ monomolecular layer (ML, 2.83 Å): in this way we control the artificially introduced disorder completely. We investigate a class of d-SL's having only few different well widths ($N_W = 4$) occurring with the same probability $P(L_{Wi})=1/4 \ \forall i\in\{1,2,3,4\}$. The barrier width and $L_{W\min}$ were 9 ML. One o-SL (sample A with $\Delta L_W = 0$ ML) and four d-SL's with different disorder strengths (samples B, C, D, and E, with $\Delta L_W = 5$, 18, 20, and 23 ML, respectively) were made. Further details on the sample preparation were reported elsewhere.[7]

The dependence of the PL spectra on T was investigated using a He closed-cycle cryostat, whose temperature could be varied from 8 to 300 K within 0.5 K of uncertainty. The samples were photoexcited by using the 514-nm line of an Ar-ion laser, whose light beam was focused on a spot of about 100-μm diameter, and the signal was detected by a cooled GaAs photomultiplier and recorded using photon counting techniques. The typical laser excitation intensity used was about 20 W/cm^2. The PL spectra at 2 K were measured on samples immersed in superfluid He. In this case the samples were photoexcited by the 647-nm line of a Kr-ion laser, and the emitted light was detected by a cooled

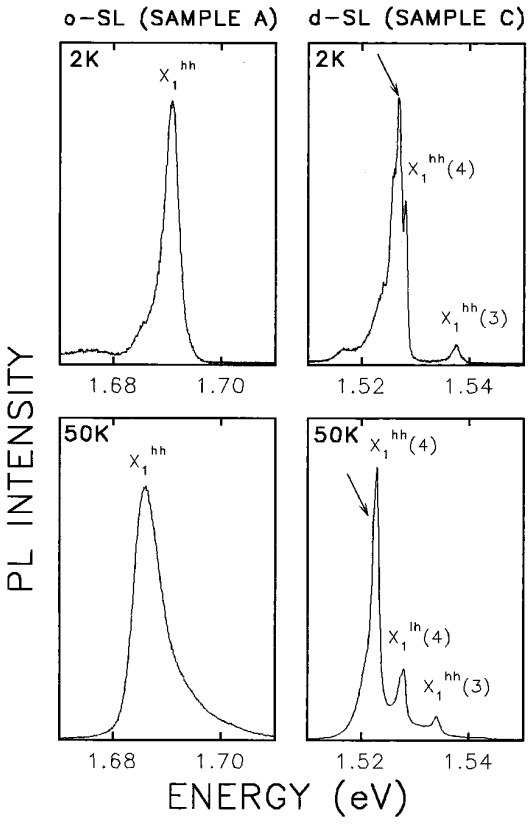

FIG. 1. PL spectra of an o-SL and a d-SL, at temperatures of 2 and 50 K. The redshift of the PL lines with T is evident. $X_1^{hh}(i)$ and $X_1^{lh}(i)$ indicate, respectively, heavy-hole and light-hole ground-state excitonic recombinations in the well of width L_{Wi}. The arrows indicate the disorder-induced fine structure.

optical multichannel analyzer. In all the PL measurements the incident laser beam was at an angle of $\pi/4$ with respect to the normal to the sample surface, and the emitted light was collected at $\pi/2$ with respect to the incident beam, and was analyzed by a double-grating spectrometer with a typical resolution of 2 meV.

We employ the matrix propagation technique[8] to obtain a quantitative description of the effect of the disorder on electronic states in the SL's. Further details on the transfer-matrix method and on the material parameters are reported elsewhere.[7] The numerical analysis shows that the introduction of disorder has two main effects: (I) the miniband containing extended states if $\Delta L_W = 0$ splits[2] into N_W subminibands of more or less localized states once $\Delta L_W \neq 0$. The splitting is more evident in strongly d-SL's. (II) These subminibands show a disorder-induced fine structure,[7] which is by contrast more evident in weakly d-SL's. In particular, we observe that the states in the subminibands at lower energy are more localized than in the highest one, where they are almost plane waves. As in the case of bulk systems containing impurities (introduced for example by doping) or amorphous semiconductors, the disorder introduces localized states below a band of extended states.

In Fig. 1 the PL spectra of an o-SL and a d-SL at two different temperatures (2 and 50 K) are compared. In the ordered case a redshift of the main emission peak of the PL spectrum with T is evident. The main emission peak X_1^{hh} is

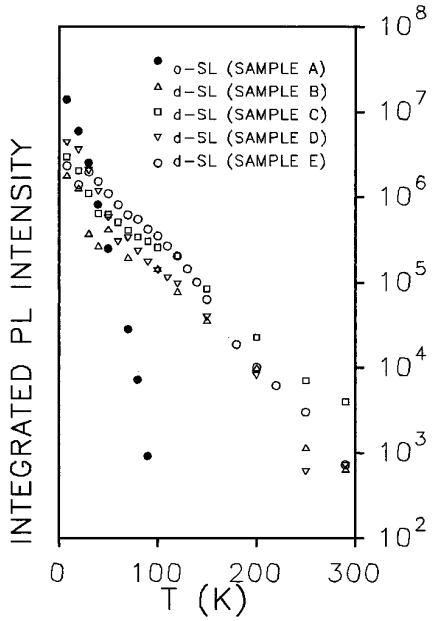

FIG. 2. Total PL intensity vs temperature for o-SL's (filled circles) and d-SL's (hollow symbols).

due[9] to radiative recombination of the ground-state heavy-hole (hh) free exciton (X). The structures on the low-energy tail of the X_1^{hh} line at 2 K are probably due to the presence of residual variations in the layer thickness, and/or to bound X. These peaks are weak, and were observed to disappear as T was increased, and to saturate as the excitation intensity I_o was increased. This well-known saturation effect is related to the relatively small number of these low-energy states in the o-SL's.

The PL emission of d-SL's is shifted to lower energies with respect to that of the o-SL's, as expected and already observed in previous studies.[4,7] As T is increased, a redshift of the whole PL spectrum is observed, as in the case of the o-SL's. Furthermore, the excitonic transitions at higher energy become more and more probable as the temperature increases. This enhancement of the high-energy recombination lines with T reflects the increasing thermal energy of the electron-hole pairs. In the spectrum of the d-SL's at $T = 2$ K, we note that the $X_1^{lh}(4)$ peak [50 meV above $X_1^{hh}(4)$] is absent, because it cannot be populated at very low T. However, the $X_1^{hh}(3)$ peak, higher in energy, is visible. This confirms the localized nature of the $X_1^{hh}(3)$ states in our disordered structures, which at low T prevents relaxation between the states in different wells (i.e., L_3 and L_4). Finally, we mention that the arrows in Fig. 1 indicate the disorder-induced fine structure, while the shoulders at lower energies are probably due to the presence of impurities and/or uncontrolled well width fluctuations, as in the o-SL: the intensity of these lines was observed to saturate as the excitation intensity I_o was increased, and some of them shifted to blue with rising I_o, indicating that donor-acceptor transitions might take place.

The integrated PL intensity at different temperatures for ordered and disordered SL's are reported in Fig. 2. While at low T the PL intensity of the o-SL's is somewhat higher than that of the d-SL's, at 100 K the PL efficiency of the

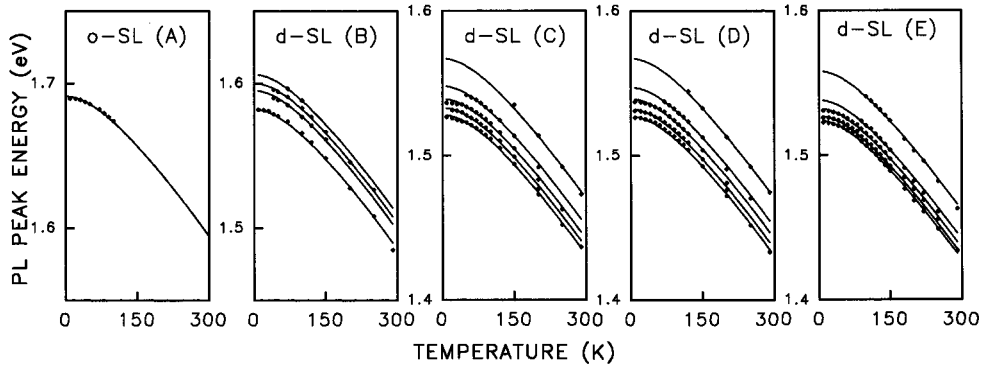

FIG. 3. The energies of the PL peaks of ordered and disordered SL's as a function of T. The observed redshift of the lines with increasing T has been fitted to the semiempirical Varshni relation (continuous lines) for the energy gap $E_g(T)$ in GaAs.

d-SL's is about three orders of magnitude higher than that of the o-SL's. Such an enhancement of the PL performance of d-SL's was already observed in short period d-SL's;[4] here we show that the enhancement of the PL efficiency can also be achieved in d-SL's having a relatively wide mean period and fully controlled disorder. This effect is a consequence of the disorder-induced localization of X along the growth axis of the samples: the localization reduces the excitonic motility, thus decreasing the nonradiative recombination probability and permitting excitonic recombination to be detected even at room temperature.

The energies of the PL peaks as a function of T for the ordered and disordered SL's are reported in Fig. 3. We remark that contradictory results were reported on the T dependence of the energies of the various PL lines: a redshift with increasing T that depends on the considered PL peak was reported by Sasaki, Wang, and Wakahara,[4] while a very weak dependence on T was observed by Arent *et al.*[5] By contrast, our results reveal a redshift of all the PL lines with increasing T. The shift as a function of temperature of all the PL features is observed to be the same. The observed shift was found to be equal to the reduction ΔE_g of the energy gap, as described by the Varshni semiempirical relationship:[10]

$$\Delta E_g(T) \equiv E_g(T) - E_g(0) = -\frac{\alpha T^2}{(\beta + T)}. \qquad (2)$$

Here $\Delta E_g(T)$ is the reduction of the gap with respect to its $T = 0$ value, and α and β are material-dependent parameters.[7] The good agreement between the experimental data and Eq. (2) (continuous lines in Fig. 3) confirms that all the PL lines of the o-SL and d-SL shift to lower energies as T increases, in the same way as the GaAs band gap does.

The T dependence of the PL spectra of the d-SL's was studied in detail by fitting the experimental spectra to a superposition of Lorentzian lines. The dependencies on T of the PL intensities of the different excitonic emissions shown in Fig. 4 have different behaviors in the low- and high-T ranges. In the limit of low T, the PL intensity of the excitonic emissions at high energy grows as T increases. In this limit, X propagation is expected to occur through variable range hopping between localized states,[2] thus as T is increased the PL lines of the localized states at higher energies are enhanced. In the high-T limit, a steeper decrease of the intensity of all the PL lines is observed. The semilogarithmic plot of the PL intensity versus $1/T$ is linear, thus indicating an Arrenhius dependence of the PL intensity, i.e., the presence of a quasithermal equilibrium. The observed activation energies $\Delta\epsilon$, obtained by fitting the data to $\exp(-\Delta\epsilon/kT)$ are, within experimental uncertainty, in good agreement with the calculated energy differences between the quasi-conduction-band of extended states (conduction subminiband $i = 1$) and the localized levels. As already observed by Chomette *et al.*,[2] the activation is governed by the electronic states, since the hole states are more weakly bound than the electron ones.

The PL measurements as a function of the excitation intensity I_o were performed at $T = 2$ K. There are no relevant differences in this dependence between ordered and disordered SL's, thus indicating a low influence of the disorder. A linear dependence over six decades is observed. The experimental investigation revealed that the main difference between the I_o dependence of the PL spectra of an o-SL and that of a d-SL is related to the behavior of the disorder-induced fine-structure recombination lines. The relative PL

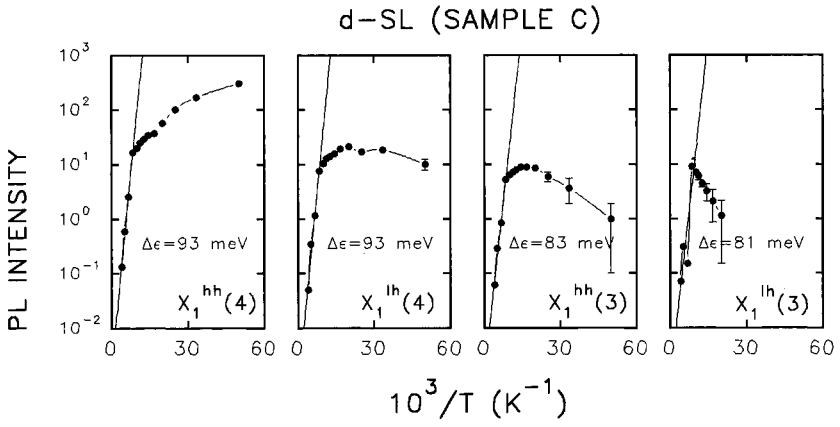

FIG. 4. Dependencies on T of the PL intensities of the various excitonic lines of a d-SL. The intensities of the PL lines were obtained by fitting the PL spectra. In the low-T limit, propagation through variable range hopping is observed. In the high-T limit an activationlike behavior is evident, and the activation energies $\Delta\epsilon$, obtained by fitting the data (continuous lines), are reported. $X_1^{\mathrm{hh}}(i)$ and $X_1^{\mathrm{lh}}(i)$ have the same meanings as in Fig. 1.

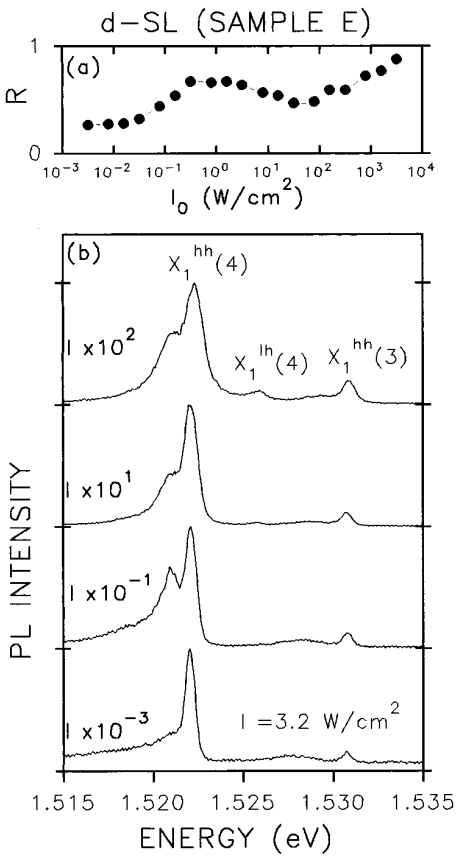

FIG. 5. (a) The intensity ratio R between the low- and high-energy parts of the PL emitted by the $X_1^{hh}(4)$ subminiband, as a function of excitation intensity I_o. The observed nonmonotonic behavior is probably connected to the strong localization of the low-energy X states (see text). (b) PL spectra of a d-SL at different excitation intensities I_o measured on the same spot. $X_1^{hh}(i)$ and $X_1^{lh}(i)$ have the same meanings as in Fig. 1.

intensities of the different lines in the disorder-induced fine structure changed as the excitation intensity was increased, showing a quite surprising nonmonotonic behavior (see Fig. 5). The low-T PL emission of the $X_1^{hh}(4)$ subminiband shown in Fig. 5(b) divides into essentially two parts, and in order to evidence the effect, the ratio R between the PL intensities of the low- and high-energy parts is reported as a function of I_o in Fig. 5(a). This nonmonotonic dependence of R on I_o can be related to the different localization properties of the states belonging to the disorder-induced fine structure. The X's created by the incident light are mainly located near the illuminated surface of the samples. At low excitation intensity I_o, the localized lower-energy states will be preferentially occupied, thus R will be relatively high. As I_o is increased, these low-energy states will be completely occupied, leading to an increase of the population of the states at higher energy and thus to an enhancement of the PL emission at higher energy, which in turn causes R to decrease. At a certain limit, I_o will be strong enough that the photoexcited carriers also start to populate the more extended states which are energetically close to the center of the subminiband. A further increase of I_o will then create carriers that can move deeper into the crystal, where they relax to the low -ying still-unoccupied states. This effect causes R to rise again. The dependence of the PL on I_o thus indicated that the states in the disorder-induced fine structure are more localized near the band edge, thus supporting our numerical results once more.

In conclusion, the PL efficiency observed at high temperatures in d-SL's is a clear experimental indication of the existence of the localized or ''impuritylike'' states predicted by various numerical studies made using the effective-mass approximation[7] as well as the pseudopotential method.[6] The investigation of the PL spectra as a function of T and I_o further confirms this picture; in particular, the measured activation energies $\Delta\epsilon$ are in good agreement with our numerical predictions. In contrast with previously reported results,[4,5] we find that all the observed PL lines shift equally to the red with increasing T, and that the red shift is equal to the reduction $\Delta E_g(T)$ of the GaAs band gap [Eq. (1)]. Finally, the relative PL intensities of the disorder-induced fine-structure lines, measured as a function of I_o, show a nonmonotonic behavior related to the differences in the localization properties of the states in the subminibands.

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