Controlling the optical properties of disordered GaAs/$Al_xGa_{1-x}As$ superlattices

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(Received 13 November 2003; published 23 March 2004)

A systematic investigation on the optical properties of disordered GaAs/AlGaAs superlattices using photoluminescence measurements was carried out. Using samples with different disorder levels we were able to show that the photoluminescence emission can be controlled by specifying the disorder of the system. In addition, the results were in good agreement with electrical measurements showing that samples having strong disorder show localization of charges but still present extended states. A qualitative theoretical model based on the effective mass approximation fits well the observed shift of photoluminescence peaks as a function of disorder.

DOI: 10.1103/PhysRevB.69.113310 PACS number(s): 73.20.−r, 73.61.−r, 78.30.Fs, 78.55.−m

I. INTRODUCTION

The electron transport and the optical properties of solids are strongly modified in the presence of disorder. While the extended states effectively contribute to the charge-transfer processes in perfectly ordered or weakly disordered crystals, the strong disorder should result in localization of carriers. Doped semiconductors where disorder is produced by a random distribution of impurities are the most used disordered electron system when studies on the effects of localization/delocalization are conducted. However, doping also increases the electron concentration and the Coulombic effects due to electron-electron interaction contribute to the disorder of the system.[1,2] The separation between these two effects can be achieved in intentionally disordered doped superlattices where both, the disorder strength and the electron concentration can be independently controlled during the growth of the samples.

The controlled disorder (while the doping level is kept constant) can be used as a tool in order to shed light on important physics principles such as the total electron localization in disordered one-dimension systems at low temperatures or the weak localization in correlated systems.[3,4] Beyond these fundamental aspects, these systems present some properties that make them important candidates for the construction of new electronic and optoelectronic devices. The disorder has a central role here: for example, in amorphous materials it is known that the high level of disorder introduce localized electronic states in the energy band gap and as a result, the observed photoluminescence in these materials are expected to be very strong. It was suggested recently that introducing sufficiently high structural disorder it was possible to tune the photoluminescence emission (PL) in ABO_3 perovskites.[5] This possibility is very promising for the development of new optoelectronic applications or for basic investigations on physics. However, it is a hard task to use the natural disorder as a control parameter in experiments.

As it is well known in semiconductor physics, during the growth of semiconductor superlattices, unavoidable random monolayer fluctuations are expected to produce disorder and charge localization, in a similar way that occur in amorphous materials. The molecular-beam epitaxy (MBE) is a technique that allows us to control all stages, atomic layer by atomic layer, of the superlattices growing. This characteristic makes MBE grown superlattices ideal systems to the investigation of effects of disorder because we can introduce artificially modifications in well/barrier width or doping, leading to the desired controlled disorder.

The new and interesting aspect of this work is the systematic study of the influence of disorder on the localized and extended superlattice states. We have investigated the optical properties of artificially disordered superlattices using low-temperature photoluminescence measurements. The level of disorder was characterized by a theoretical parameter defined by the energy ratio between the Gaussian distribution of the electron energy of an isolated quantum well and the miniband width of the nominal superlattice in the absence of disorder. We show that is possible to control the intensity and energy of the emission using different levels of disorder in the samples. The experimental results for the position of PL peaks were in agreement with the theoretical ones. Furthermore, capacitance-voltage experiments have shown unambiguously that extended and localized states coexist even in the most disordered samples.

II. EXPERIMENT

The samples used here are silicon doped $(GaAs)_m(Al_{0.3}Ga_{0.6}As)_6$ superlattices (SL's) and they were grown by MBE on (100) oriented GaAs substrates. In order to avoid the short-range in-plane fluctuations, the growth of the superlattices was interrupted for 20 s. at the normal interface and for 3–5 s. at the inverted one. The total number of 50 periods was grown for each structure. The vertical disorder was introduced by a controlled random variation of the GaAs well thicknesses around the nominal value $m=17$ monolayers, corresponding to a Gaussian distribution of the lowest levels of noninteracting electrons forming the conduction miniband. The barrier thicknesses and the uniform doping level (6×10^{17} cm^{-3}) were kept unchanged in all the samples.

According to the calculations made by the Kronig-Penney model including the potential nonparabolicity, the width of the lowest Γ miniband of the nominal superlattice $(GaAs)_{17}(Al_{0.3}Ga_{0.6}As)_6$ is $W=55$ meV.[4] The disorder strength was uniquely characterized by the disorder param-

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eter $\delta=\Delta/W$, where Δ is the full width at half maximum of a Gaussian distribution of the electron energy calculated in a isolated quantum well and W is the miniband width of the nominal superlattice in the absence of disorder. Even in the nominal superlattices the unavoidable monolayer fluctuations produce a vertical disorder strength $\delta\approx0.18$. One expects that at $\delta\simeq1$ majority of the electrons moving in the miniband perpendicular to the layers should be localized. The localization of the vertically moving electrons was detected in the same superlattices by Raman scattering in Ref. 6.

Electrical (Schottky capacitance) measurements were used in order to characterize the disorder effects on the charge distribution. For the Schottky diode construction, we fabricated a conventional Ohmic contact to the substrate using an AuGeNi alloy annealed at 450 °C for 120 s; the Schottky contact was formed by the deposition of a 200 nm gold layer with 500 μm diameter. The capacitance was measured using a standard lock-in technique (with a SR530 Stanford dual-phase lock-in amplifier). The samples were mounted in a helium closed cycle cryostat and the capacitance-voltage curves were obtained at 80 K.

PL experiments were taken using a U1000 Jobin-Yvon double grating spectrometer coupled to a cooled GaAs photomultiplier and a conventional photon counting system. The 457.9 nm line of an Ar^+ ion laser was used as exciting wavelength, with the laser's maximum output power kept at 200 W/cm^2. A cylindrical lens was used to prevent the overheating of the samples. The slit width used was 100 μm and all measurements were taken at 80 K.

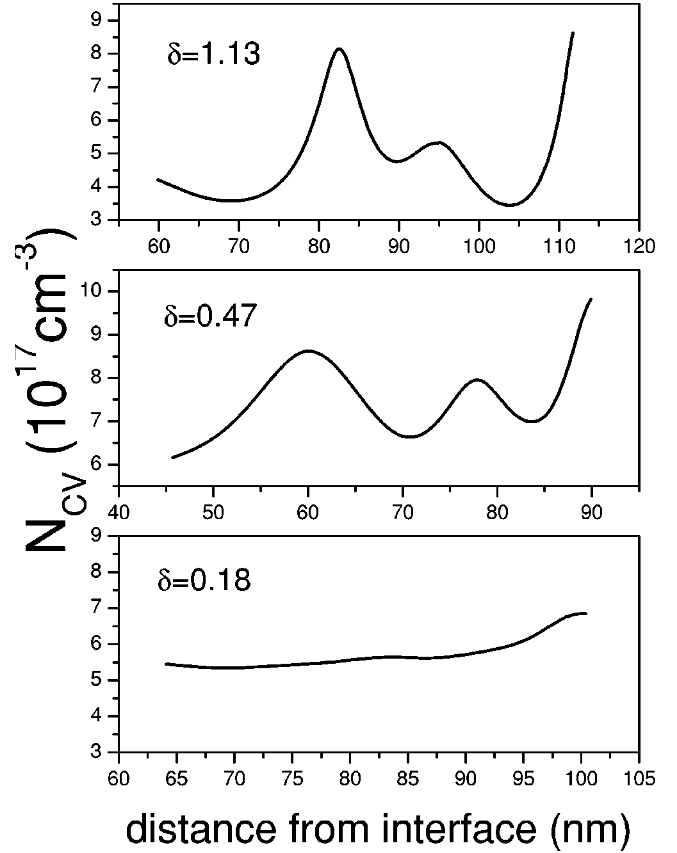

FIG. 1. *C-V* profiles of the superlattices with different levels of disorder taken at 80 K.

III. RESULTS AND DISCUSSION

Before conducting the optical studies, the influence of the intentional structural disorder on the electronic properties of the superlattices under investigation was studied using *C-V* measurements. The disorder introduced during the growth of the samples by the variation of the well thicknesses produces a modulation of the coherent band state energies of the superlattice, spreading them into a random Gaussian-like distribution with a characteristic width Δ. As a consequence, it induced a random modulation of the local electron density. Using *C-V* measurements, the spatial distribution of the electrons along the growth direction $[N_{C-V}(z)]$ was calculated by the usual equations for the *C-V* profile,[7]

$$N_{C-V}(z)=\frac{-2}{qS^2\varepsilon_s}\left[\frac{dC^{-2}}{dV}\right]^{-1}, \tag{1}$$

and

$$z=\frac{\varepsilon_s S}{C}, \tag{2}$$

where C is the measured capacitance, S is the Schottky device area, and the other symbols have their usual meanings.[8] The effect of potential modulation is shown in Fig. 1, where the *C-V* profiles of the superlattices with different degrees of disorder were plotted. These profiles were obtained at $T=80$ K and the spatial resolution of the *C-V* measurements is given by the screening length which in this case is about 5 nm. As it can be seen, a much larger variation of the local electron density was found in the superlattice with the stronger disorder although in all the samples the average electron concentrations were found in a good agreement with the nominal doping concentration. This observation seems to be an indication that extended and localized states are coexisting. We should not expected all the electrons to be localized in our structures because these superlattices cannot be considered as one-dimensional systems. In fact, they are three-dimensional structures as revealed by previous Shubnikov-de Haas measurements:[4,9] the vertical disorder (on the growth direction) have influenced the transport properties on the parallel direction. It should be noted that these extended states are not Bloch-like but they represent the coupling between adjacent energy levels where the coherence length of the structure was unchanged.

The PL spectra of the samples presenting different levels of disorder are shown in Fig. 2. The first point to be noted is the PL intensity evolution of the spectra. The intensity increases about one order of magnitude from the ordered to the most disordered SL. This enhancement can be explained by the changes of the band structure induced by the increasing disorder and is currently attributed to carrier localization effects associated with the disorder.[10–13] This is an indication that using disorder we can control some of optical properties of the superlattices and the observed dependence of the PL intensity with the disorder level can be used in new technological applications.

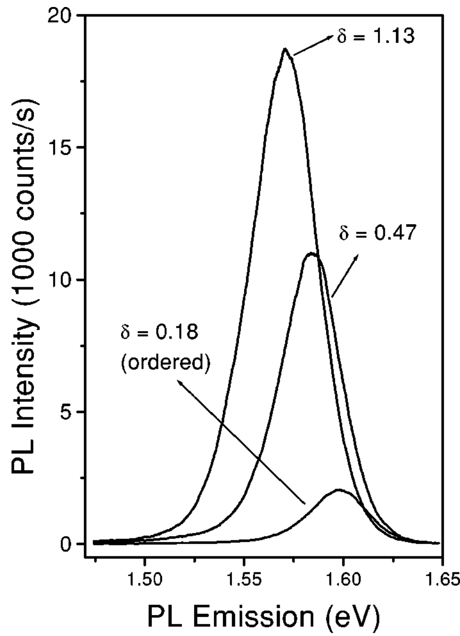

FIG. 2. Evolution of PL spectra for samples presenting three different disorder levels. These measurements were taken at 80 K.

More interesting discussion can be done using the analysis of the variation of the Stokes shifts observed in Fig. 2. Some authors have already studied and classified the origin of such shifts considering localization effects in disordered SL's.[10,14] Referring to the PL peak of a GaAs well ($W1$) as a probe, Chomette *et al.* have investigated extended states, vertical transport, and carrier localization effects in GaAs/AlGaAs weakly disordered SL's. They correlated the reduction of the $W1$ photoluminescence intensity to the creation of localized levels. In parallel to this reduction it can be observed a redshift of the PL peak for increasing disorder. Bellani *et al.* have investigated the existence of delocalized states in random dimer SL's.[3] In their work, a theoretical calculation was employed as a base to attribute the blueshift of the experimental PL peak to the occurrence of localized states as an effect of the disorder.

In our results, the observed redshift of the PL band was caused by the increase of the disorder. PL experiments contains not enough information to distinguish the agent responsible by the shifts, i.e., PL can not distinguish the extended and localized electron states. It can only define an electronic transition through an energy gap. So, confronting this observation with C-V measurements we believe the redshift should be, in fact, correlated to the presence of localized states. In the absence of disorder the band structure is organized in energy levels distributed in minibands and the PL emission should be attributed to the recombination of electrons and holes in the gap [see Fig. 3(a)]. On the other hand, the intentional disorder is responsible by the localization of electrons presenting energies around the energy of the miniband. The C-V results have demonstrated that even samples having strong disorder such as $\delta = 1.13$ still present extended states, explaining the observed redshift. It should be noted that the induced Gaussian disorder sprays the energy levels around the ordered miniband energy, but only those ones

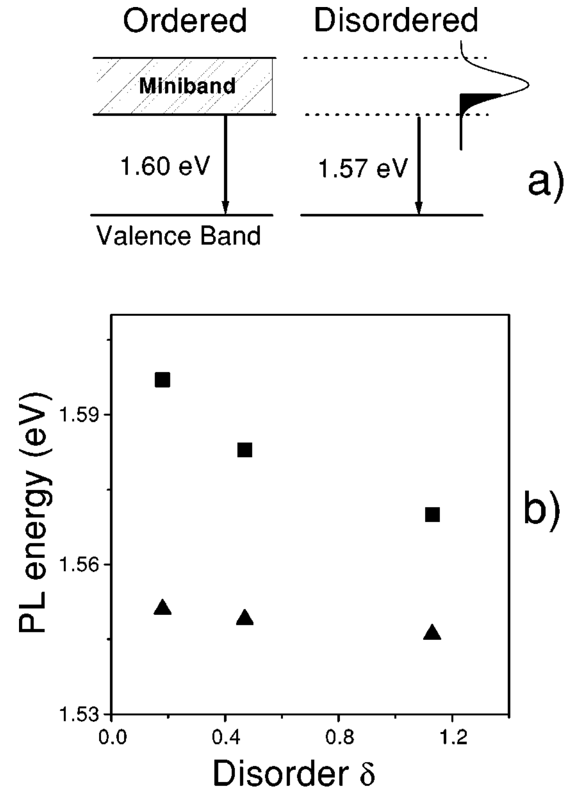

FIG. 3. (a) Simplified energy-band diagram of the ordered and of the most disordered superlattices. The Gaussian curve represents the calculated dispersion of the electron levels of the disordered SL and the black area represents the levels that effectively contribute to the PL emission; (b) experimental (squares) and theoretical (triangles) PL energy shifts.

with lower energy (with respect to the energy miniband minimum) will be able to give a strong contribution to the PL process. Using this idea, increasing the disorder level, both the intensity and the redshift increase as well, in agreement with the experimental results.

In addition, we applied a theoretical model in order to account the effects of disorder in the electronic structure of the superlattices and then calculate the allowed band to band transitions. The model was based on the effective mass approximation using only structural inputs such as doping levels and layer thicknesses as parameters. The Schrödinger and Poisson equations were solved in a self-consistent procedure. First, the electric potential along the growth direction [$\phi(z)$] was supposed to be given only by the conduction-band discontinuities. Next, starting with this initial function for the potential, the eigenvalues (E) and the wave functions [$\psi(z)$] were obtained, using the Schrödinger equation

$$\left[-\frac{\hbar^2}{2m^*}\frac{d^2}{dz^2} + V(z) \right] \psi(z) = E\psi(z), \tag{3}$$

where

$$V(z) = -q\phi(z) + \Delta E_C(z), \tag{4}$$

and ΔE_C is the band discontinuity at the GaAs/AlGaAs interfaces. Then, the electron distribution along the structure is

calculated and the Poisson equation determines a new potential $\phi(z)$ and consequently a new $V(z)$ which, in turn, is used in Eq. (3) to update the eigenvalues and wave functions. The process stops when the difference between the potentials $[V(z)]$ is lower than 10^{-6} eV. All donors were considered to be ionized.

The results are depicted in Fig. 3(b) and they agree very well with the experimental ones: as the disorder increases, the PL peaks are redshifted. Figure 3(b) emphasizes the sensibility of the Stokes shift to the disorder variation. The difference between the observed and calculated energies was due to simplifications on the model: the system was considered as an one-dimensional structure (z direction) and the calculations do not take into account holes quantization. The Gaussian profile of the disorder was responsible for the linear variation of the PL energies. This allows one to infer about the possibility of tuning the energy of the light emission in semiconductor devices. In this case, considering the *state of the art* of the MBE growth process in semiconductors, these materials reveal certain advantages in the device industry in comparison to other material groups as the ABO_3 perovskites, for example.

IV. CONCLUSION

Photoluminescence properties of disordered GaAs/AlGaAs superlattices were investigated as a function of an artificially structural disorder induced by random variation of the wells width. The investigation of the optical properties of samples growth with different level of disorder was focused on the possibility of tunning the PL energy and intensity by controlling the electronic spectra of the samples. The results showed that the induced Gaussian disorder leads to a linear variation of the PL energy which in turn, points to a possible new technological application. The effects of the structural disorder was also studied by capacitance-voltage measurements, which indicate that extended and localized electronic states coexist even in the most disordered samples.

ACKNOWLEDGMENTS

The authors would like to acknowledge Professor Yu. A. Pusep for the samples used here. This work was partially supported by the Brazilian agencies CNPq, FAPESP, and CAPES.

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