

Electronic structure and vertical transport in random dimer GaAs-Al_xGa_{1-x}As superlattices

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We report a systematic study of several GaAs-Al_xGa_{1-x}As semiconductor superlattices grown by molecular-beam epitaxy specifically designed to explore the existence of extended states in random dimer superlattices. We have confirmed our previous results [V. Bellani *et al.*, Phys. Rev. Lett. **82**, 2159 (1999)] with much additional evidence that allows us to lay claim to a clear-cut experimental verification of the presence of extended states in random dimer superlattices due to the short-range correlations (dimers) that inhibit the localization effects of the disorder.

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I. INTRODUCTION

Spatial overlap between electronic states of neighboring quantum wells (QW's) in a semiconductor superlattice (SL) shifts the degenerate energy levels and leads to the appearance of minibands separated by minigaps, as was predicted long ago by Esaki and Tsu.¹ In their seminal paper, these authors speculated that a periodic modulation of the composition of a semiconductor at a length scale smaller than the electron mean free path would result in the occurrence of negative differential conductance.² Furthermore, if the length of the SL is shorter than the mean free path, coherent transmission through an ideal SL is to be expected.³ Subsequent advances in semiconductor research made it possible to firmly establish these predictions on solid experimental grounds.⁴

Successful experimental validation of former predictions arising from purely theoretical investigation suggests that SL's are ideal candidates to carry out research projects in basic physics. Advances achieved in molecular-beam epitaxy, which allow the production of high-quality SL's tailored with the desired conduction- and valence-band profiles, support the feasibility of this belief. In recent years, there have been remarkable examples of this appealing trend in solid state physics. More than 70 years ago, Bloch considered the motion of electron wave packets in crystals subject to an external applied electric field.⁵ Work by Bloch was further clarified and elaborated by Zener,⁶ who pointed out that an electron that is not subjected to scattering processes will perform an oscillatory motion, the so-called Bloch oscillations. Clear evidence of such oscillations was reported by Feldmann *et al.*⁷ and Leo *et al.*⁸ Moreover, 40 years ago it was proposed that a Stark-Wannier ladder should appear in periodic solids subject to an applied electrical field⁹ although

it could not be observed until the end of the 1980s, after the advent of SL's.¹⁰

More recently, intentionally disordered SL's, in which disordering is artificially created by random thickness fluctuations, have received much attention.¹¹⁻³⁰ These intentionally disordered SL's were grown with the main aim of observing Anderson localization and mobility edges. The reported enhancement of photoluminescence (PL) was attributed to electron and hole localization due to disorder. Evidence of the existence of extended states in impuritylike SL's (namely, two ordered SL's joined by a wide QW) were reported by Lorusso *et al.*³¹ It is thus apparent that semiconductor SL's are suitable systems for controllable experiments on localization or delocalization and related electronic properties.

An ideal semiconductor superlattice forms a one-dimensional (1D) structure in the growth direction that has been used successfully to study theoretically or experimentally the electronic properties of 1D systems.^{32,33} In this context, some authors proposed that SL's are *physically realizable systems* that allow for a clear-cut validation of the existence of extended states in low-dimensional random systems with correlated disorder.³⁴⁻³⁷ These systems are characterized by the key ingredient that structural disorder is short-range *correlated*. A number of tight-binding³⁸⁻⁴⁰ and continuous⁴¹ models of correlated disordered 1D systems predicted theoretically at the beginning of the 1990s the existence of extended states, in contrast to the earlier belief that *all* the eigenstates are localized in 1D disordered systems. But, owing to the lack of experimental confirmation, there was some controversy as to the relevance of these results and their implications for transport properties. Recently, our group presented clear *experimental evidence* of this phenomenon in semiconductor SL's.⁴² To be specific, the experimen-

tal PL energies were in very good agreement with the calculated electronic states,³⁶ suggesting the formation of delocalized extended states. Most important, vertical dc resistance of both correlated disordered SL and ordered SL were very similar, showing no temperature dependence at low temperature, as should be expected for transport in the presence of extended states. These measurements led us to conclude that Anderson localization is inhibited in correlated disordered SL's.

In this work, we report further progress along the lines given in the preceding paragraph. We provide further evidence of the existence of extended states in correlated disordered SL's, and we study carefully the physical nature of such states as well as their effects on optical and transport phenomena. The paper is organized as follows. In Sec. II we present our model and summarize our previous theoretical work,^{34–37} which we find convenient for a better understanding of the present paper, specifically as regards the behavior of the transmission coefficient and dc conductance. To check the relevance of theoretical predictions, we grew three different types of GaAs-Al_{0.35}Ga_{0.65}As SL's, namely ordered SL (OSL), random SL (RSL) without spatial correlation, and random dimer SL (DSL) with dimerlike correlation. Section III describes these samples in detail as well as their x-ray characterization. In Sec. IV we present PL spectra at low temperature from the various SL's. This allows us to perform the analysis of the experimental transition energies and the ascertainment of the localization and delocalization properties of the SL's from comparison with calculations.³⁶ The body of the paper is Sec. V where we present an extensive discussion of transport properties of the three types of SL's determined by a variety of techniques: dc conductivity versus temperature, *I-V* characteristic, photovoltage (PV), and short circuit photocurrent (PC). Finally, in Sec. VI, we discuss our results and, from the comparison between transport properties of RSL and DSL, we conclude that Anderson localization is inhibited when correlations are intentionally introduced in the sample.

II. MODEL AND BACKGROUND

We summarize in this section previous results of ours³⁶ for correlated disordered GaAs-Al_{*x*}Ga_{1–*x*}As SL's, which will be useful for the discussion of optical and transport properties, addressed in the next sections. For our present purposes, it is enough to focus on electron states close to the band gap with $\mathbf{k}_\perp = \mathbf{0}$ and use the one-band effective-mass framework to calculate the envelope function. The electronic states were calculated using a Kronig-Penney model that has been shown to hold in the range of well and barrier thicknesses we used.⁴⁵ Let us assume d_w and d_b to be the width of QW's (GaAs layers) and barriers (Al_{*x*}Ga_{1–*x*}As layers), respectively. The thickness of barriers separating neighboring QW's is assumed to be the same in the whole SL, $d_b = b$. OSL's are constructed by also assuming the thickness of QW's to be the same in the whole SL, $d_w = a$. In our model of RSL, we consider that d_w takes at random one of two values, a and a' . Finally, a DSL is built by imposing the additional constraint that QW's of thickness a' appear only

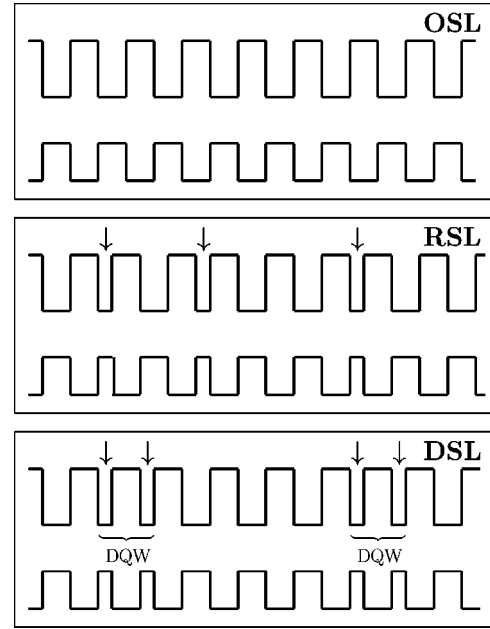


FIG. 1. Schematic diagram of the conduction- and valence-band profiles of the three SL's considered in this work. Arrows indicate the center of narrow QW's for clarity.

in pairs, called hereafter a dimer QW (DQW), as shown in Fig. 1.

We now consider a single DQW in an otherwise ordered SL. The condition for an electron to move in the OSL is given by³⁴

$$\left| \cos(\kappa a) \cosh(\eta b) - \frac{\kappa^2 - \eta^2}{2\kappa\eta} \sin(\kappa a) \sinh(\eta b) \right| \leq 1, \quad (1)$$

where $\kappa^2 = 2m^*E/\hbar^2$ and $\eta^2 = 2m^*(\Delta E_c - E)/\hbar^2$. Here ΔE_c is the conduction-band offset. The origin of energies is taken at the GaAs conduction-band edge. We have taken a constant effective mass m^* at the Γ valley, but this is not a serious limitation as our description can be easily generalized to include two different effective masses. For the growth parameters corresponding to the SL's used in the present work (see Sec. III), there is only one allowed miniband below the barrier, ranging from 0.1 up to 0.2 eV. Any electron moving in the SL will be reflected back at the DQW except if its energy matches a resonant energy E_r obtained from the following condition³⁴

$$\cos(\kappa a') \cosh(\eta b) - \frac{\kappa^2 - \eta^2}{2\kappa\eta} \sin(\kappa a') \sinh(\eta b) = 0. \quad (2)$$

In our DSL this energy is $E_r = 0.15$ eV and thus it lies within the allowed miniband. This means that the reflection coefficient at the DQW vanishes and, consequently, there exists complete transparency at the resonant energy E_r . In the vicinity of the resonance, the reflection coefficient is nonzero but rather small. Choosing a' appropriately is important in allowing us to locate the resonant energy E_r within an allowed miniband of the periodic SL, that is, the resonant

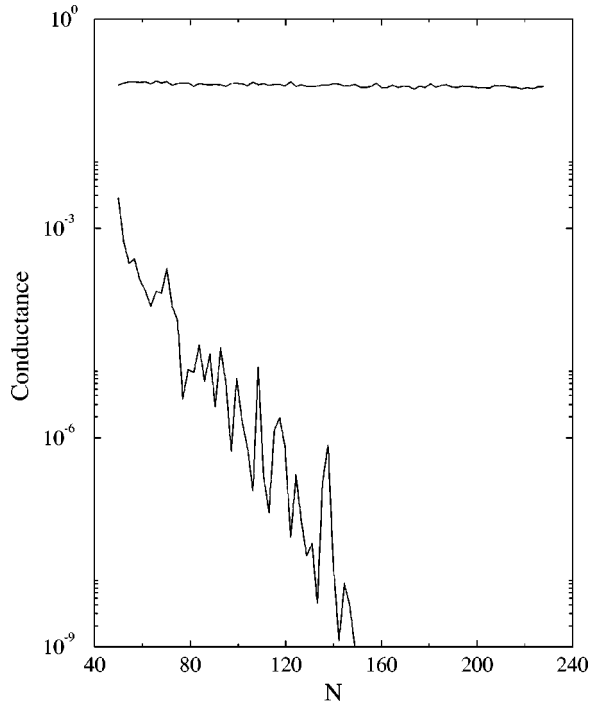


FIG. 2. dc conductance, in units of e^2/h , at 77 K as a function of the number N of barriers in intentionally disordered GaAs-Al_{0.35}Ga_{0.65}As SL's when the chemical potential is 0.15 eV, i.e., it matches the resonant energy. Upper (lower) curve corresponds to DSL (RSL). We present results of averages over 100 different SL's for each case.

energy in the range of energies given by Eq. (1). When several DQW's are introduced at random to build up a DSL, the transmission coefficient is still unity at the resonant energy, as can be easily demonstrated,³⁴ and the corresponding state is delocalized in spite of the fact that the SL is disordered. Close to this resonance there are a number of states whose localization length is larger than the system size and, consequently, they behave like extended ones as regards transport properties.

We now turn to the problem of transport across disordered SL's. The extended or localized nature of electronic states close to the Fermi level can be elucidated from the dependence of the dc conductance on the number of layers in the SL. The states are extended (localized) when the dc conductance is constant (decays exponentially) as the SL size increases, thus leading to an Ohmic (non-Ohmic) behavior of the sample. In Fig. 2 we can see the dependence of the dc conductance at 77 K on the number of barriers when the Fermi level matches the resonant energy E_r for both RSL and DSL. Notice that the behavior is Ohmic only for the DSL. We have obtained the dc conductance through the following expression, earlier discussed in detail by Engquist and Anderson:⁴⁶

$$\mathcal{G}(T, \mu) = \frac{e^2}{h} \frac{\int \left(-\frac{\partial f}{\partial E} \right) \tau(E) dE}{\int \left(-\frac{\partial f}{\partial E} \right) [1 - \tau(E)] dE}, \quad (3)$$

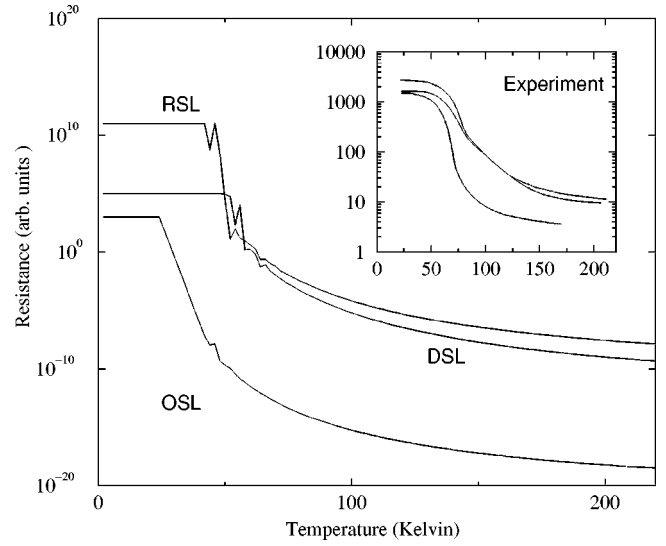


FIG. 3. Computed temperature dependence of the vertical resistance for the RSL, DSL, and OSL. In the inset are shown the experimental measurements for the three SL's in the same order.

where integrations are extended over the allowed bands, τ is the transmission coefficient, f is the Fermi-Dirac distribution, and μ denotes the chemical potential of the sample. We also studied the temperature dependence of the transport properties. The current density can be calculated within the stationary-state model^{47,48} by the expression

$$j(V) = \frac{m^* e k_B T}{2 \pi^2 \hbar^3} \int_0^\infty \tau(E, V) N(E, V) dE, \quad (4)$$

where T is the temperature, V the applied bias, and k_B the Boltzmann constant; the electron transport through the SL is described through the resonant tunneling mechanism. $N(E, V, T)$ accounts for the occupation of states on both sides of the device, according to the Fermi distribution function, and it is given by

$$N(E, V) = \ln \left(\frac{1 + \exp[(E_F - E)/k_B T]}{1 + \exp[(E_F - E - eV)/k_B T]} \right), \quad (5)$$

E_F being the Fermi level. In this framework we have calculated the dependence of the resistance on the temperature for a *constant electric potential* of 0.1 V. The data are plotted in Fig. 3. We compare the numerical results with our previous measurements,⁴² which are shown in the inset. There is a reasonable qualitative agreement with the experimental values for the three types of SL's. We can note that by lowering the temperature, both the calculated and experimental resistances increase and, at the lower temperatures, the curves flatten, in agreement with experiments by other authors.⁴³ This behavior can be roughly understood by considering that as the temperature decreases, the broadening in energy of the Fermi-Dirac distribution function around the Fermi level shrinks, by making more and more restrictive the conditions for the electron tunnelling between adjacent wells. However, when this broadening becomes comparable with the distance

between the intraminiband levels of the SL, the current becomes temperature independent. We will come back to this point later.

III. SAMPLES AND X-RAY CHARACTERIZATION

Now let us turn the attention to our experimental results and how they compare with theory. To this end, we grew several GaAs-Al_{0.35}Ga_{0.65}As SL's, and we studied their electronic properties by PL at low temperature and dc vertical transport. The samples are three undoped SL's grown by molecular-beam epitaxy. All the SL's have 200 periods and Al_{0.35}Ga_{0.65}As barriers 3.2 nm thick. The conduction-band offset is $\Delta E_c = 0.25$ eV and the effective mass is $m^* = 0.067m$, m being the electron mass. In the OSL, all the 200 wells are identical with a thickness of 3.2 nm. In the RSL, 58 wells are replaced by wells of a thickness of 2.6 nm, and this replacement is done randomly. The DSL is identical to the RSL, with the additional constraint that the wells of a thickness of 2.6 nm appear only in pairs.³⁶ In the latter sample, the disorder exhibits the desired short-range spatial correlations. In each sample, the SL is cladded on each side by 100 nm of *n*-type Al_{0.3}Ga_{0.7}As, Si doped to 4×10^{18} cm⁻³, with a 50 nm *n*-type GaAs buffer layer (doped to 4×10^{18} cm⁻³) on the substrate and a 3 nm *n*-type GaAs cap layer (doped to 6×10^{18} cm⁻³).

We measured x-ray diffraction spectra of the SL's with a double-crystal diffractometer, in order to check their structural parameters. This technique has been shown to be very powerful for the study of periodicity and disorder in semiconductor superlattices.^{44,49,50} The diffraction curve at (004) symmetric reflections for the two disordered samples show satellite peaks of the order of ± 1 lying close to $\pm 0.8^\circ$ with respect to the GaAs peak. These satellite peaks are located at identical positions for the two disordered SL's, showing that the random SL's have identical periods. Therefore, the dimer constraint intentionally introduced during sample growth is the only difference between RSL and DSL.

IV. PHOTOLUMINESCENCE CHARACTERIZATION

PL has proved to be a good technique in the study of the electronic properties of SL's,^{11,12} giving transition energies between confined electron states. PL spectra were taken as a function of the temperature in the 4–300 K range (light power density on the sample of about 2.5 W/cm²). The exciting source was an argon laser ($\lambda = 514.5$ nm); the light emitted by the samples was analyzed by means of a 0.5 m Jobin Yvon-Spex HR 460 single monochromator and detected by a cooled PbS detector through a conventional lock-in technique. The spectral resolution was better than 1 meV.

Figure 4 shows the PL spectra at low temperatures of the three SL's; in the inset, a sketch of the radiative transitions in the three SL's is drawn. The temperature dependence of the energy of the near-band edge peaks is different for the three samples, but the energy shift between them is almost independent of temperature on a wide range. The low-temperature PL peak for the OSL, which lies at 1.69 eV, is

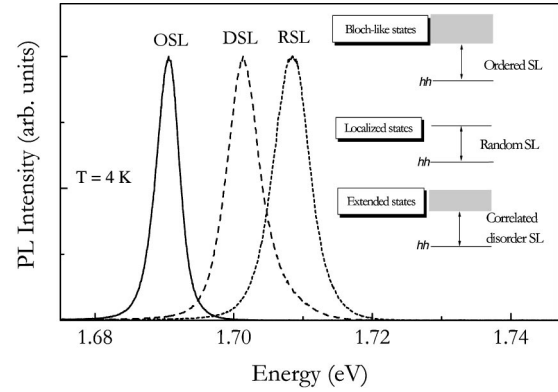


FIG. 4. PL spectra at $T=4$ K of the OSL, DSL, and RSL.

due to recombination between electrons in the conduction band and heavy holes in the valence band. The energy of this transition is in good agreement with the calculation of the miniband structure performed using a Kronig-Penney model, the calculated lower energy of the miniband being very close to the energy at which PL intensity rises. The PL peak of the RSL is at higher energies with respect to the other two samples. In this SL, the intentional disorder introduced by the random distribution of wells 2.6 nm thick localizes the electronic states.³⁶ The energy blueshift is of 20 meV with respect to the OSL and compares well with the calculations of the transition energy, assuming that the exciton binding energy is the same in the SL's.

The PL peak of the DSL is close to 1.70 eV and, as can be clearly seen in Fig. 4, redshifted with respect to the PL peak for the RSL. According to Fujiwara⁴⁵ this redshift is due to the formation of a miniband with a tunnel process for carriers between the GaAs wells. This result strongly supports previous theoretical predictions of the occurrence of a band of extended states in correlated disordered SL's. The values of the transition energies agree with the values predicted by our models,⁴² indicating that the predictions were fundamentally correct.

V. TRANSPORT MEASUREMENTS

PV at open-circuit and short-circuit PC experiments were performed in the temperature range between 10 and 80 K. These techniques have been used widely to investigate semiconductor quantum heterostructures.^{30,51–59} The light source was a white light produced by a halogen lamp passed through a monochromator, with a spectral resolution of 2 meV. The spectra were measured in the photon energy range between 1.5 eV and 2.1 eV. The photoelectrical signal was revealed by using a lock-in technique. The spectral response of the optical system was measured in the whole range and used to normalize the spectra.

The PV spectrum taken at $T=10$ K confirms the PL results. Moreover, practically coincident spectra were obtained by measuring the short-circuit PC signal. In fact, for each sample the excitonic peaks are at the same energies as in PL spectra, with a slight difference that could be due to a Stokes shift. It is worth noting that generally a photovoltaic or a photocurrent signal is the result of the light modulation of

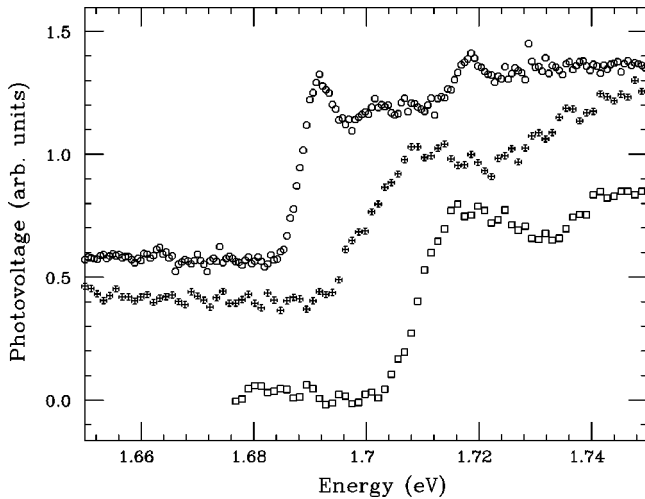


FIG. 5. Photovoltage spectra of the OSL (circles), DSL (crosses), and RSL (squares) at $T=30$ K.

the barrier potential or the collection of the photogenerated minority carriers, both requiring the presence of an inner junction. Moreover, as confirmed by recent experimental results,^{55,59} the energetic position of the peaks depends on the optical absorption process, whereas their intensity and line shape are also related to the transport mechanism and therefore, indirectly, to the potential profile along the direction of the electric field. In the present case, we measured an open-circuit PV signal despite the absence of any intentional junction. In effect, in our structure low (weak) potential barriers are present between the n^+ and n regions. As proof of this fact we observed a clear correlation between the intensity of the PV signal and a significant asymmetry of forward and reverse I - V characteristics taken at each temperature: in other words, the photovoltaic effect rises for an unintentional non-Ohmic behavior of the n - i - n structure. At temperatures above 80 K the nonlinearity of the I - V curves disappears, and the possibility to monitor the PV signal at higher temperatures is prevented. In addition, the lowest-energy peak in the PV spectrum appears to be better resolved when the asymmetry of the I - V curve is maximum, and this happens at a temperature of ≈ 30 K, even if the PV signal shows a strong monotonic reduction with the temperature increase. Figure 5 shows the PV spectra of the SL's at this temperature. The above observation also suggests that the temperature evolution of the peaks is dominated by uncontrolled factors such as the accidental presence of an internal electric field, in addition to the generation rate and the transport mechanism of the carriers. Therefore, in the present case a line shape analysis of the peaks becomes unreliable. On the contrary, the significant information is given by their position.

I - V characteristics were taken at different temperatures in the range between 10 K and 300 K. The curves were taken by setting the current values and measuring the steady-state voltages in a two-point geometry. The measurements were performed in the dark, by increasing the temperature from 10 K to room temperature, after the sample was illuminated at the lowest temperature with white light. This latter procedure

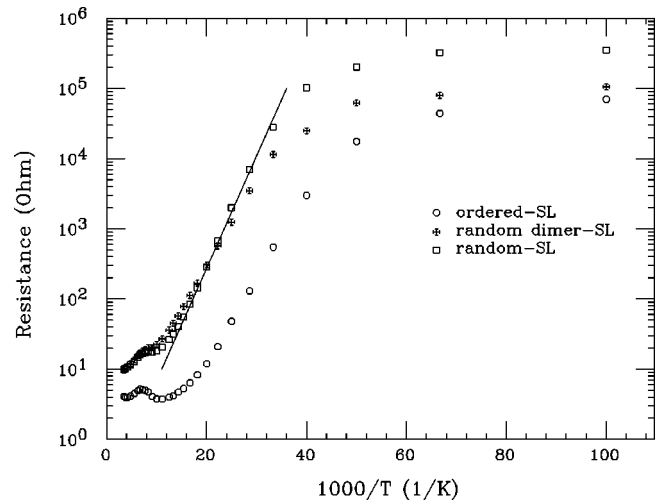


FIG. 6. Arrhenius plot of the experimental electrical resistance of the OSL, DSL, and RSL. The diameter of the mesa structure is $160 \mu\text{m}$ in the case of the OSL and $200 \mu\text{m}$ in both the other cases. The interpolation in the region of linearity is reported for the RSL.

proved useful to contrast the reduction of the injection efficiency of carriers from the n^+ -doped contact layers into the SL structure due to the electron trapping into the DX centers: this deep level is introduced in the n -type $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ material by the donor atoms themselves.^{60,61} Owing to the thermally activated electron capture cross section of the DX center, the photoionization of the latter at a low temperature actually increases persistently the free electron density in the conduction band until it reaches a saturated value (persistent photoconductivity effect). The electron capture into the DX centers becomes appreciable above ≈ 70 K; the equilibrium occupancy of the deep level is restored above ≈ 150 K.

The sample resistance was calculated as a function of the temperature, $R(T)$, from the linear region of I - V curves. Since at low temperatures ($T \leq 100$ K) the linear regime is restricted to a very narrow range and the curves tend to become nonsymmetrical with respect to $V=0$, in these conditions the $R(T)$ values were obtained by averaging the slopes of the forward and reverse I - V data around zero. In order to compare $R(T)$ values taken in the same way, and in a nearly Ohmic regime, the calculation of this quantity was performed at a very low field (1–10 V/cm). The resistance data thus obtained are slightly different from those we presented recently⁴² (see the inset of Fig. 3) since in that work we measured $R(T)$ by applying a constant current of 1 mA and reading the voltage drop across the sample, without illumination.

Figure 6 reports the “Ohmic” resistance of the SL's versus $1000/T$. The qualitative behavior of the $R(T)$ curves agrees with the results of Fig. 3; in particular, we can observe that the resistance of the OSL is systematically lower than the other two at all the temperatures, as expected. In addition, the Arrhenius plot of the data reveals a possible regime of activated transport. When the I - V curves were taken without the low-temperature photoionization of the DX centers, a behavior on the whole similar to the one shown in Fig. 6 was obtained for $R(T)$, but with higher

absolute values of the sample resistance; also, the narrowing of the linear range of the I - V curves at a low temperature and their nonsymmetry were in any case observed. This result confirms that the low-temperature photoionization of the DX centers does not modify the probability of crossing the SL region by the carriers, but it reduces the ‘‘external resistance’’ in series to it (R_{ext}) and, above all, restores persistently the highest free carrier density that can be supplied by the contact layers.

Returning to Fig. 6, four temperature regions can be distinguished.

(1) *High temperatures* ($150\text{ K} \leq T \leq 300\text{ K}$). The DX center occupancy is at equilibrium, and the SL resistance is in series with the resistance of the contacts, which is low but appreciable; in addition, a contribution due to the external circuit (bonding, wires, etc.) cannot be excluded.

(2) *Intermediate temperature* ($70\text{ K} \leq T \leq 150\text{ K}$). The data are scarcely meaningful because the density of the free carriers supplied by the n -doped $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layers, and also the I - V curves, evolves in time, owing to the activation of the process of electron capture into DX centers. This phenomenon is more and more evident as the temperature increases, and it is responsible for the weak shoulder observed in the $R(T)$ curves in this range.

(3) *Middle-low temperatures* ($25\text{ K} \leq T \leq 70\text{ K}$). For all the samples the resistance shows an activated regime, the activation energies increasing from the OSL ($\approx 20\text{ meV}$) and DSL ($\approx 23\text{ meV}$) to the RSL ($\approx 29\text{ meV}$). As regards the transport mechanism responsible for such an activated regime, (i) the thermoionic emission over the barriers can be excluded as a dominant mechanism, because the SL quantum level (or miniband) in the well is over 100 meV below the barrier top; (ii) the phonon-assisted hopping could be considered; (iii) moreover, it can be observed that the SL regions are $\approx 1\ \mu\text{m}$ thick, and therefore a bowing of the structure can be expected in all the SL’s; (iv) also, the spike in the conduction-band profile at the interface with the n -doped $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layers could play a significant role. However, we notice that in this range the absolute value of the OSL resistance is close to an order of magnitude lower than that for the other samples, and we can observe that the activation energy increases from the OSL, DSL, to the RSL: this tentatively points to the relating of the region of activated transport to the SL structure itself (more than to R_{ext}). This point will be further investigated in the future.

(4) *Low temperatures* ($10\text{ K} \leq T \leq 25\text{ K}$). The $R(T)$ curves are weakly dependent on temperature, as in the case

of transport by tunneling.^{43,62} In this temperature region, the resistance of the DSL approaches the OSL, in agreement with our expectations.

VI. CONCLUSIONS

We studied the structural, optical, and transport properties of ordered and intentionally disordered superlattices with and without correlation of the disorder. It was found that the introduction of a short-range correlation in a disordered SL leads to the formation of extended states, theoretically expected. The PL experiment shows that the correlation of the disorder in a semiconductor SL leads to the delocalization of the electronic states, with tunneling of the electrons and redshift of the electronic transition energies. The values of the electrical resistances at a low temperature shows that the RSL has the highest values of resistance, while the OSL and the DSL have comparable values, confirming the formation of bands of extended states in the DSL. In an intermediate-temperature range ($25\text{ K} \leq T \leq 70\text{ K}$) the SL’s show a regime of activated transport, with close values of the activation energies for the OSL and DSL; the RDL has the highest value.

During this presentation we discussed the misleading definition generally accepted for the localization theorem. In fact, while it is undoubtedly true that in the low-dimensional systems the presence of unintentional or random (i.e., uncorrelated) disorder induces localization, it has now been established that this conclusion is not obvious when the disorder presents some kind of correlation. The possibility to have extended states in disordered systems with special characteristics as a function of the degree and kind of disorder opens up an attractive *scenario* concerning fundamental physics speculations. Moreover, the possibility to *control* the localization effects by taking advantage of the enormous possibilities offered by the advanced growth techniques and nanotechnologies opens the way to a wide field of applications towards devices based on new concepts.

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