Resonant Rayleigh scattering in ordered and disordered semiconductor superlattices

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We report the experimental study of resonant Rayleigh scattering in GaAs-AlGaAs superlattices with ordered and intentionally disordered potential profiles (correlated and uncorrelated) in the growth direction z. We show that the intentional disorder along z modifies markedly the energy dispersion of the dephasing rates of the excitons. The application of an external magnetic field in the same direction allows us to modify the exciton localization and to study the relative modification of the exciton dephasing, energies, and linewidth.

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INTRODUCTION

In recent years, a large number of studies have been devoted in exploring the general properties of disordered systems through the investigation of the optical response and transport properties of semiconductor nanostructures. An important topic in the physics of disordered systems is the influence of an intentionally disordered potential profile along the growth axis z in semiconductor superlattices (SL's) on quantum interference and carrier localization.[1–3] SL's are excellent candidates in performing such a study since the advances achieved in molecular beam epitaxy (MBE) allow the fabrication of SL's tailored with desired conduction- and valence-band profiles. In this way, it is possible to design a great variety of one dimensional potentials with intentional disorder in the growth direction z (vertical disorder) which coexists with the in-plane disorder due to unintentional growth islands. The global result is an anisotropic disorder which effect on the quantum interference has become an interesting subject of recent experimental studies. As an example, magnetotransport measurements in doped semiconductor superlattices recently allowed the study of the effects of the anisotropy on the phase-breaking time of the electrons.[2]

In the present work, we measure simultaneously the continuous wave resonant Rayleigh scattering (RRS), the photoluminescence (PL), and photoluminescence excitation (PLE) spectra of semiconductor SL's in a magnetic field to study the influence of the vertically correlated and uncorrelated anisotropic disorders on the coherence relaxation and exciton localization. We use this technique since RRS has access to information on the effective localization and on the spectral dispersion of the dephasing,[4] while the PL and PLE spectra in magnetic field are valid techniques to study the exciton delocalization along z.[5] At the same time, the external magnetic field allows one to continuously tune the in-plane exciton localization, with respect to the vertical one.

RRS is the resonant light scattering into all directions due to the localization of the exciton center of mass wave functions, and is proved to be a powerful tool for the study of dephasing process mainly in quantum wells (QW's)[6–15] and also in microcavities,[16] quantum wires,[17] and bulk semiconductors.[11,18] Rayleigh scattering in semiconductors arises from imperfection breaking the symmetry of translational invariance of the crystal due to imperfection in the bulk, on the surface, or from interfaces. However, crystals of very high quality can be grown by means of MBE, and in these crystals the defect scattering can be extremely low. In such systems and, in particular, in GaAs-AlGaAs multiple quantum wells (MQW's), Hegarty *et al.* observed for the fist time a strong contribution to the Rayleigh scattering in the vicinity of an optical resonance.[6,7] In their pioneering experiment, the authors observed that RRS was enhanced 200-fold with respect to the non resonant surface and defect scattering. This RRS arises from the spatial fluctuation in the local resonant frequency which, because of the strong dispersion near resonance, leads to corresponding fluctuations in the refractive index. Hegarty *et al.* also showed that RRS is a valid experimental technique to study the localization and/or delocalization of two dimensional excitons in QW's.[7] Despite the analogy between PL and RRS, these two phenomena have completely different natures. PL is an incoherent process due to the relaxation of a real exciton population. On the other hand, RRS derives from the coherent scattering of light as a consequence of the relaxation of wave vector in a real solid and mainly originates from localized exciton states.[9,11,14]

Experimental spectroscopic techniques such as time resolved RRS and RRS spectral speckle analysis have been recently used with success in order to extract quantitative information on structural disorder from RRS (see, for instance, Ref. 4 and references therein). In particular, the measured spectrally and time resolved RRS spectra have been analyzed with accurate models which calculate the RRS tak-

ing into account all the important features of semiconductor heterointerface fabrication. Specifically, Savona and Langbein used either a time propagation of the excitonic polarization with a phenomenological dephasing or a full excitonic eigenstate model including microscopically calculated dephasing rates and phonon scattering rates.[4] The experimental spectra well agree with the calculations, demonstrating the role of many elements of the disorder affecting the RRS.[4,19] Between these elements, we have the interface structure, correlations, and anisotropy.

EXPERIMENT

In this paper, we study the RRS, PL, and PLE in $Al_{0.35}Ga_{0.65}As/GaAs$ ordered, intentionally disordered, and correlated disordered SL in an external magnetic field up to 14 T. The excitation light was supplied by a Ti-sapphire laser pumped by an Ar^+-ion laser. The excitation laser light was polarized σ^+ and the component σ^+ of the scattered and emitted light has been detected. The samples are three n^+-i-n^+ heterostructures with a SL grown by MBE in the intrinsic region, grown by MBE. All the three samples have a SL of 200 periods and $Al_{0.35}Ga_{0.65}As$ barriers 3.2 nm wide. In the ordered SL (OSL), all the 200 wells are identical with well width of 3.2 nm (hereafter referred to as A wells). In the random SL (RSL), it contains 142 A wells and 58 wells of thickness 2.6 nm (hereafter referred to as B wells) and these B wells appear randomly. The so-called random dimer SL (DSL) is identical to the RSL with the additional constraint that the B wells appear only in pairs. In the latter sample, the disorder exhibits the desired short-range spatial correlations. Additional details on the samples can be found in previous publications.[3,20] Our previous theoretical and experimental results[3,20,21] showed that whereas in the RSL the states are localized, the DSL supports a narrow band of critical (non-Bloch-like) extended states, while the OSL supports a wide band of Bloch extended states. Therefore, in the OSL the carriers are mobile either in the growth direction or in the plane. On the other hand, for RSL and DSL we found that the mobility is intimately related to the disorder and that the exciton dynamics is affected by the presence of correlations that completely modify the localization of the electronic states in the growth direction.[3,21]

RESONANT RAYLEIGH SCATTERING AND PHOTOLUMINESCENCE EXCITATION

In Fig. 1, we report a series of emission spectra (luminescence plus scattered light) measures on the (a) OSL, (b) DSL, and (c) RSL, scanning the energy of the excitation laser light, for several hundred different values of the excitation energy for polarized excitation and detection. These emission spectra have been plotted using small dots, and two main structures can be identified, one with high density of points corresponding to PL and the other with less dense point corresponding to RRS. For clarity, in the inset of the upper panel of Fig. 1(a), we show only 19 of these spectra (for which the excitation energy differs by 0.8 meV from one spectrum to the other). When the laser energy is set off resonance, we observe a weak Rayleigh scattering due to defect and imperfections. On the other hand, when the laser energy is set close to the resonance, the Rayleigh scattering increases dramatically and reaches a peak some tenth times the off-resonance defect scattering. In the same figure, we include the PLE spectra measured detecting at the energy of the maximum of the PL peak. The PLE spectrum of the OSL [Fig. 1(a)] shows a main peak due to the heavy hole exciton (HH) and a smaller one due to the light hole one (LH). This attribution of the LH transition is in agreement with the recent work of Gerlovin *et al.* who measured PLE spectra in SL samples with similar well and barrier widths.[22] As regards the DSL, its PLE spectrum [Fig. 1(b)] shows of a main peak at 1.698 eV due to the transition between the valence and conduction minibands which are present due to the dimer-type correlation of the disorder given by the paired 2.6 nm wells. The spectrum also presents a wider structure at 1.707 eV which is due to excitonic transitions in the 3.2 nm

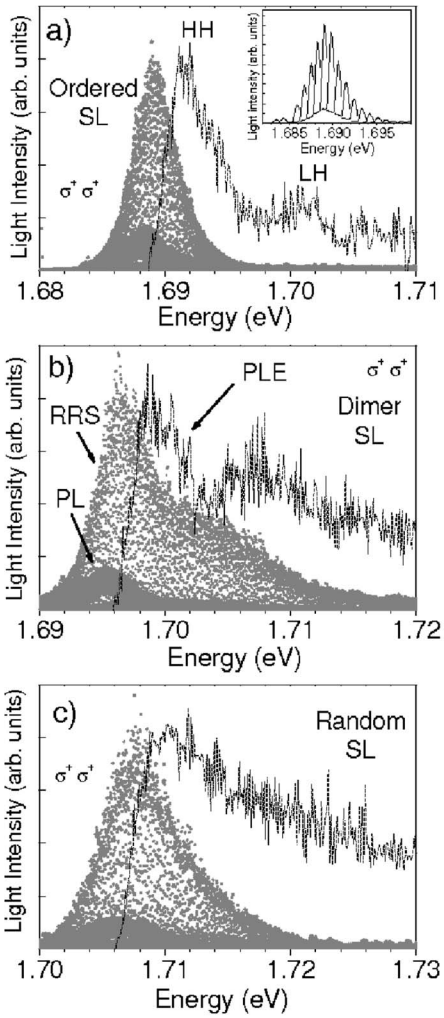

FIG. 1. Scattered light spectra (small points) and PLE spectra (dashed line) of the (a) ordered SL, (b) random SL, and (c) random dimer SL at 4 K and zero external magnetic field for σ^+ excitation and detection polarization. In the inset of panel (a), we show only some of these spectra (for which the excitation energy differs by 0.8 meV from one spectrum to the other).

isolated wells. These two structures are also observed in the PL spectrum and in the RRS since the latter is a scattering phenomenon resonant with the PL. Finally, for the RSL [Fig. 1(c)] the PLE and the RRS spectra of this sample at zero magnetic field show a main structure with a high energy exponential tail typical of the disordered systems.

The PL and RRS spectra of the three SL's are clearly different due to their different electronic structures. In particular, they are wider in the two disordered SL's due to their intentional disorder which affects directly the PL linewidth. In particular, the full width at half maximum (FWHM) of the PL and of the RRS at zero magnetic field is 4.2 meV in the OSL, 6.8 meV in the RSL, and 5.7 meV in the DSL. Also, the RRS shows similar behavior, with FWHM of 3.9, 7.6, and 6.4 meV in the OSL, RSL, and DSL, respectively, evidencing the resonant contribution of the PL to the Rayleigh scattering in the vicinity of the inhomogeneously broadened optical resonance.

As has been shown by Fujiwara,[30] the increasing of the delocalization of the carriers between the GaAs wells in a SL induced a redshift of the absorption edge, and therefore of the PL. In our DSL, we observed this redshift of the PL with respect to the RSL and this was interpreted with the increase of delocalization of the carrier and confirmed trough vertical transport experiment.[3] This increased delocalization of the carriers with respect to the RSL, due to the correlation of the disorder, is also reflected in a reduction of the PL linewidth.

As regards the analysis of the RRS, Hegarty *et al.*[6] in their pioneering experimental study in semiconductor QW's interpreted the experiments with the presence of a mobility edge to explain the dispersion of the dephasing time. Their work was described theoretically by Takagahara who studied in detail the link between the energy dependence of the homogeneous linewidth and the dispersion of the dephasing time.[23] More recent theories and experiments show that the RRS can be interpreted without the need of introducing a mobility edge.[4,24] In particular, Savona and Langbein developed very accurate theoretical calculations of the RRS using exciton eigenstates and microscopically calculated dephasing rates due to radiative decay and phonon scattering, which are in very good agreement with the experiments.[4]

The observation of a shift between RRS and PLE in semiconductor QW's for the HH transition was interpreted as a consequence of the inhomogeneous broadening of the excitonic transition.[6–11] In contrast, in bulk GaAs and in GaAs-AlGaAs QWs intentionally grown with high amount of interfacial disorder, the shift of the RRS with respect to the PLE was found to be negligible.[11] According to the description of Hegarty *et al.*,[6] the shift is due to a rapid increase of the homogeneous linewidth $\Gamma_h(\omega)$. On the other hand, the absence of a shift between RRS and PLE can be interpreted as a constant value for $\Gamma_h(\omega)$ in the case of either localized or propagating excitons. With respect to the origin of the shift between RRS and PLE in semiconductor QW's, some authors ascribed it to the presence of a mobility edge between propagating and localized states[6,7,9] although other works report alternative explanation of this phenomenon.[4,24]

The three SL's that we study in this work have been grown in the same identical condition one after the other, and thus their in-plane disorder can be assumed to be same. The aim of our work is to show that the RRS and PLE spectra are markedly different in the three SL's and their behavior can be justified on the basis of the different exciton localization along z due to the specific SL potential profile.

We can see in Fig. 1 that the main PLE peak at zero magnetic field has a different shape and width in the three SL's due to their different electronic structures. In particular, it is wider and not symmetric in the RSL and DSL due to their intentional disorder. This makes it difficult to determine the exact position of the PLE peak in the RSL and DSL and therefore to compare quantitatively the shift between PLE and RRS in the three samples.

Therefore, in order to extract more information on the energy dispersion of the dephasing time, we calculated the energy dispersion of the homogeneous linewidth $\Gamma_h(\omega)$ using the model of Hegarty *et al.*:[6,7]

$$\Gamma_h = \left[\frac{I_s(\omega)}{1 - T^2(\omega)} + \Gamma_x^{-1} \right]^{-1} - \Gamma_0, \tag{1}$$

with $I_s(\omega)$ the backscattered intensity which is proportional to the RRS intensity, $\Gamma_x(\omega)$ the observed (predominantly inhomogeneous) linewidth, T the transmission of the sample, and Γ_0 a fix parameter related to the layer thickness and to the correlation length of the unintentional width fluctuations.

In our experiments, we did not measure the absolute value of optical absorption since it requires the etching of the substrate with equipments not available and also because we performed the optical experiments using a setup with optical fibers for the excitation and detection of the light, which does not allow the easy measurement of the optical absorption.

Instead, we measured the PLE spectrum which is anyway proportional to the optical absorption (being indeed also called pseudoabsoption). In order to obtain the optical transmission T, we firstly normalized to unity the maximum of the PLE for the three samples and we assumed an equal reflectivity in the three samples in the 5 meV energy range where we calculate $\Gamma_h(\omega)$. This approximation is also justified by the fact that we want to compare the relative $\Gamma_h(\omega)$ between the three SL's and not to get the absolute values. Therefore, using a constant reflectivity, taken as zero for simplicity, we get T as 1 minus the PLE value. The normalization to 1 of the maximum of the PLE corresponds to considering a transmission equal to zero at the energy of the maximum of the PLE, i.e., total optical absorption, which is sound for 200 period SL's at the energy above just above the exciton absorption edge. We also normalized to unity the RRS spectra of the three SL's.

Equation (1) is only accurate when $\Gamma_0 \ll \Gamma_h \ll \Gamma_x$, and as Hegarty *et al.* showed, this is valid in experiment in two dimensional heterostructures.[6,7] Therefore, we can disregard $1/\Gamma_x$ and Γ_o because they do not modify the energy dependence of $\Gamma_h(\omega)$ (only change slightly its absolute value) and rewrite Eq. (1) as

$$\Gamma_h \approx \left[\frac{I_s(\omega)}{1 - T^2(\omega)} \right]^{-1}. \tag{2}$$

In Fig. 2, we report the homogeneous linewidth for the three SL's calculated using Eq. (2) from the experimental

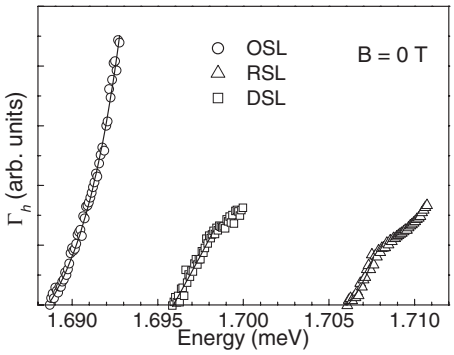

FIG. 2. Energy dispersion of the homogeneous linewidth for the three SL's at zero magnetic field. The solid lines are best fits using exponential functions.

data of Fig. 1. We can see that the $\Gamma_h(\omega)$ for the OSL increases monotonously with a behavior similar to that of ordered MQW's studied by Hegarty *et al.*[6,7] On the other hand, in the two disordered SL's the homogeneous linewidth also shows initially a fast increase but exhibits a shoulder at energies around 1.698 and 1.708 eV in the DSL and RSL, respectively. This behavior in the disordered SL's derives from the fact that the RRS is not symmetric but shows some small structures at energies just above its maxima (see Fig. 1), which can be attributed to higher energy excitons. We can also note that this shoulder appears a little later in the DSL than in the RSL.

According to the description of Hegarty *et al.*[6] a rapid increase of the homogeneous linewidth is ascribed to the presence of a mobility edge between propagating and localized states. For excitons at energies below the mobility edge, the dephasing by phonon gives an exponential growth of the $\Gamma_h(\omega)$ as a function of energy [see Eq. (4) of Ref. 32],

$$\Gamma_h \propto \exp\left[\frac{E - E_m}{kT}\right], \tag{3}$$

with E_m being the energy of the mobility edge and E the energy of the exciton, and this behavior has been experimentally verified in ordered MQW's.[7] Therefore, we fitted the $\Gamma_h(\omega)$ dispersion of the OSL using an exponential function that shows good agreement with our data. For the RSL and DSL, we also obtain good agreement with an exponential growth before the shoulder appears. The best fits are the solid lines in Fig. 2. As we mentioned, the exponential behavior in the OSL holds in the entire energy range and this agrees with the work of Takagahara which showed how in an ordered quantum well heterostructure there exists a transition energy region, few meV wide, between delocalized and localized regimes[23] with a gradual change between the two regimes. Our results show how the differences in the electronic structure in the z direction affect the homogeneous linewidth in this energy region. The OSL exhibits a wide band of extended states along z and the exciton dephasing appears similar to that of ordered MWQ systems. The DSL and RSL exhibit a very different behavior, indicating that the intentional disorder along z modifies markedly the dephasing rates of the excitons.

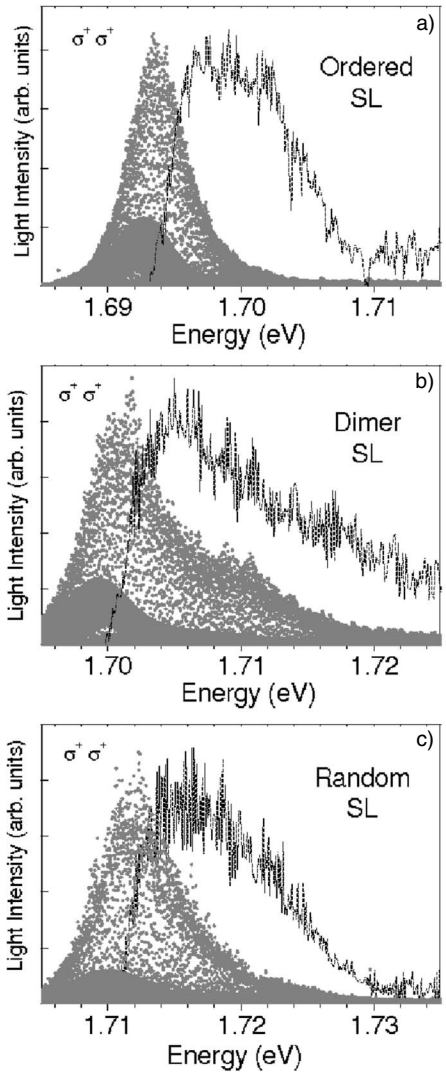

FIG. 3. Scattered light spectra (small points) and PLE spectra (dashed line) of the (a) ordered SL, (b) random SL, and (c) random dimer SL in an external magnetic field of 14 T for σ^+ excitation and detection polarization.

From Eq. (1), we could estimate the energy of the mobility edge (E_m). A reasonable criterion could be to use the energy where $\Gamma_h = 1$. This is a crude estimation but is valid in comparing the behavior of homogeneous linewidth dispersion in three SL's. For the OSL we obtain $E_m = 1.6901$ eV, for the DSL $E_m = 1.6978$ eV, and for the RSL $E_m = 1.7083$ eV. We will come back to these values after the presentation of the data with an applied magnetic field.

In Fig. 3, we report a series of emission spectra (luminescence plus scattered light) measured on the (a) OSL, (b) DSL, and (c) RSL, scanning the energy of the excitation laser light, for several hundred different values of the excitation energy as in Fig. 1 but now in the presence of a 14 T magnetic field applied in the growth direction and in Faraday configuration. We observe that by applying a magnetic field, the PLE, PL, and the RRS spectra move toward higher energies due to the diamagnetic shift of the excitons. At the

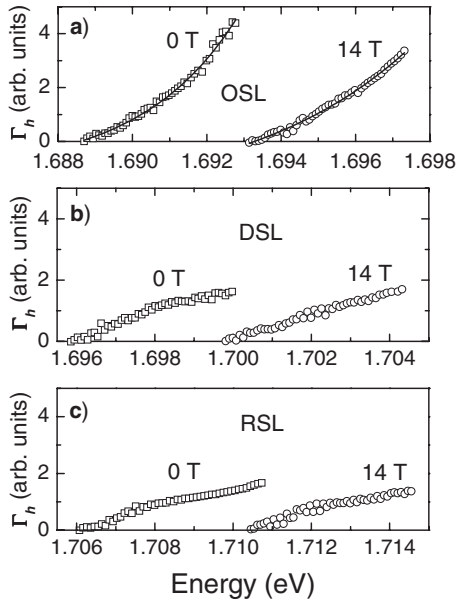

FIG. 4. Energy dispersion of the homogeneous linewidth $\Gamma_h(\omega)$ for the (a) OSL, (b) DSL, and (c) RSL at 0 and 14 T. The solid lines are best fits using an exponential function.

same time, the HH peak widens and splits into several substructures. We observe that the magnetic field broadens and splits the RRS spectra into several substructures due to the excited states of the exciton. In particular, the FWHM of the PL at 14 T is 4.8 meV in the OSL, 6.6 meV in the RSL, and 5.9 meV in the DSL. Also, the FWHM of the RRS shows a similar behavior, i.e., the value for the DSL is between that for the OSL and that for the DSL. In fact, the RRS FWHM is 4.9, 8.1, and 7.9 meV in the OSL, RSL, and DSL, respectively, showing the resonant contribution of the PL to the Rayleigh scattering in the vicinity of the inhomogeneously broadened optical resonance. Also, at 0 T the FWHM of the PL and of the RRS for the DSL was between the value for the OSL and that for the DLS, and this indicates that the magnetic field does bleach out this relation which is associated with the different electronic structures of the three SL's. In the next section, we will go back to this point.

The magnetic field also modifies the dispersion of the homogeneous linewidth. In Fig. 4, we show the homogeneous linewidth for the three SL's at 14 T, and for the sake of comparison in the same graphs we plot again the values at 0 T. We can see that for each SL, the $\Gamma_h(\omega)$ at 14 T are shifted ~5 meV to higher energy with respect to that of 0 T due to the diamagnetic shift of the excitons. For the OSL, the magnetic field reduces the rate of growth of Γ_h, indicating an increase of the exciton localization, whereas in the two disordered SL's, this effect is very weak.

Applying the same criterion used for the dispersion at 0 T, we can estimate the energy of the mobility edge E_m at 14 T. For the OSL we obtain E_m=1.6950 eV, for the DSL E_m=1.7023 eV, and for the RSL E_m=1.7128 eV. To compare our results, in Fig. 5 we plot the homogeneous linewidth at 0 and 14 T vs $E-E_m$, where E_m is the corresponding value for each curve. As discussed before, here we see clearly that for the OSL the homogeneous linewidth rise at 14 T is

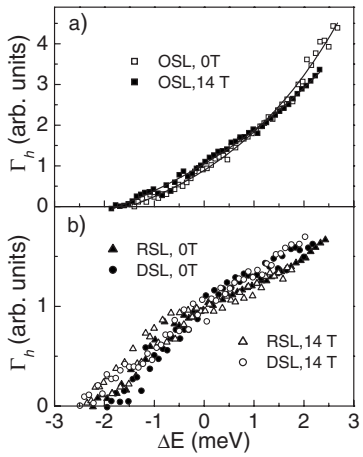

FIG. 5. $\Gamma_h(\omega)$ for the three SL's at 0 and 14 T. The curves have been shifted in energy, at ΔE=0 corresponding to Γ_h=1. The solid lines are exponential fits of the dispersion for the OSL.

smaller than at 0 T and this comportment can be ascribed to the increased carrier localization induced by the magnetic field. On the other hand, in the two disordered SL's the $\Gamma_h(\omega)$ behavior and quantitative increase are similar at 0 T up to 14 T and this can be attributed to the fact that in the RSL and DSL, the carriers are much more localized than in the OSL, and that the additional localization induced by the magnetic field is not resolved.

PHOTOLUMINESCENCE SHIFT AND LINEWIDTH

In Fig. 6, we report the diamagnetic shift of the energy of the PL as a function of the magnetic field applied in the growth direction of the SL's. The dashed line is a quadratic fit for OSL. We can observe that the energies of the main PL peak of the three samples shift to higher energy in proportion to the square of the magnetic field, as expected for excitonic transitions.[25] The shift is nearly the same for the three SL's; at this level the presence of a SL, ordered or disordered, has not a detectable effect of the exciton energy. By fitting with a

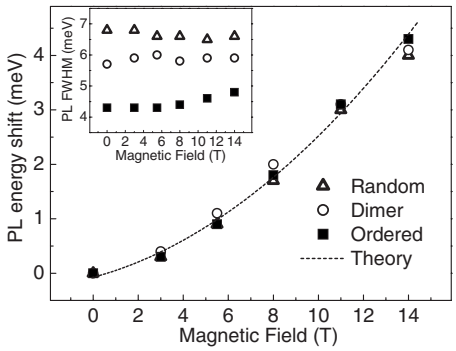

FIG. 6. Energy shift of the energy of the PL for the three SL's as a function of the magnetic field. The dashed line is a quadratic fit of the shift for the ordered SL. The inset shows the full width at half maximum (FWHM) of the PL peak as a function of the magnetic field.

quadratic expression the data for the OSL (with well and barrier widths of 3.2 nm), we find the diamagnetic shift to be 22 $\mu eV/T^2$. This value is very close to the earlier published value of 21 $\mu eV/T^2$ measured in QW with well width of 2.1 nm.[26] Our finding that the diamagnetic shift of a SL is close to that of a QW with similar well width was also observed in the past by Birkedal *et al.*[27] In their work, they observed that a SL with well and barrier widths of 8 and 5 nm had a shift of 26.4 $\mu eV/T^2$, close to the value of 29 $\mu eV/T^2$ measured on QW with 7.5 nm well width.[26] Moreover our smaller value of the diamagnetic shift with respect to the one measure by Birkedal *et al.*[27] is coherent with the observations that the diamagnetic shift increases with the increase of the well width.[25,26]

On the other hand, it is observed that the magnetic field affects differently the evolution of the emission linewidth in the three SL's. In the inset of Fig. 6, we show FWHM of the main PL peak of the three SL's as a function of the magnetic field. All the FWHMs refer to the PL measured with an excitation laser energy 0.025 eV above the PL peak position. As mentioned previously, at zero field the linewidth of the two disordered SL's is greater than that of the OSL due to the disorder which increases the predominantly inhomogeneous linewidth of the PL. Moreover, at all fields the PL is narrower in the DSL than in the RSL. This behavior, also evidenced in previous work,[3] indicated that the intentional disorder really enhanced the linewidth and that the correlation of the disorder leads to its small reduction.[28]

The linewidth of the PL of the OSL increases very slightly with the magnetic field, whereas the behavior is not monotonous in the other two SL's. The increase of the linewidth in the OSL is in agreement with recent results of a work with PL experiments on QW's in magnetic fields.[29] In this work, the increase with the magnetic field of the FWHM of the excitonic recombination in ordered QW's is attributed to the squeezing of the exciton size induced by the magnetic field, which, in turn, leads to a reduction in the spatial averaging of the potential energy fluctuations of the QW due to the finite exciton size.

On the other hand, in the RSL and DSL, the linewidth changes not monotonously with the magnetic field. It increases for fields up to 2 T in the RSL (up to 4 T in the DSL), then decreases, and increases again above 8 T. In these SL's, the additional source of disordered is the well width intentional variation in the z direction. As we mentioned previously, the three SL's have been grown in identical conditions so we can reasonably assume that the in-plane disorder is in average the same. Therefore, the decrease of the PL width at the intermediate magnetic fields is ascribable to the intentional disorder along z. At zero magnetic field, the radius of the exciton in the z direction in GaAs-$Al_{0.35}Ga_{0.65}As$ QW's with well width of $\sim$3.2 nm is $\sim$2 nm (while it is $\sim$12 nm in the plane).[31] Therefore, the exciton radius along z is comparable with the width of the wells contained in our two disordered SL's. The fact that the evolution of the PL width in the disordered SL has a behavior different from that in the OSL suggests that the magnetic field applied along z not only reduced the exciton radius in the plane but also affected the exciton extension along z. The decrease of the broadening of the PL in the 2–8 T range could indicate that the localization length and the exciton extension along z are acting competitively and that the overall result is partial delocalization of the excitons along z with the field.

In summary, we studied the RRS, PLE, and PL in ordered and intentionally disordered semiconductor SL's in the presence of an external magnetic field. From the RRS and PLE spectra, we calculated and compared the behaviors of the energy dispersions of the dephasing times, relating them to the ordered and disordered nature of the SL's. We also studied the behavior of the emission energy and linewidth with the magnetic field, comparing the results with the available results obtained on QW systems. As regards the dependence of the PL linewidth on the field, we found distinct difference in the evolution of the PL width between the ordered and disordered SL's. While in the ordered SL the PL linewidth increases monotonously with the field, in the disordered SL the evolution is not monotonous and this behavior can be correlated to the interplay between localization length and exciton extension in the z direction along which the intentional disorder is present.

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