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Spectroscopic evidence of the dissymmetry of direct and inverted interfaces in GaAs/AlAs type-II superlattices

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In type-II pseudodirect superlattices (SL's) the study of the polarization of photoluminescence in a magnetic field along the growth axis brings insight into exciton localization and interface quality. We demonstrate that excitons in a given sample are not localized at random at the direct or inverted interface, but mostly at one type of interface. For each sample we determine the nature of this interface. This determination is made possible by the study of two asymmetrical superlattices in which excitonic recombination is forced at one type of interface by the design of the band structure. In nominally symmetrical SL's, i.e., with a two-layer period, we show that localization at the direct or the inverted interface is determined by the growth conditions, namely, the substrate temperature and growth interruption at interfaces. [S0163-1829(98)03607-8]

I. INTRODUCTION

The structure of heterointerfaces in multilayered systems such as quantum wells (QW's), superlattices (SL's), and quantum wires has a strong influence on their electronic and optical properties. It has been much investigated by means of high-resolution electron transmission microscopy with chemical lattice imaging,[1,2] cross-sectional scanning tunneling microscopy,[3,4] and optical spectroscopy such as photoluminescence (PL) (Refs. 5–9) and Raman scattering.[7,10] The interface roughness is generally defined as the existence of interface islands of thickness 1–2 ML. The size and lateral spacing of these interface islands have been shown to depend on the growth conditions.[6] However, the existence of residual roughness on an atomic scale has been shown even in high-quality samples.[1,7,11] An atomic exchange process during growth is likely to be one of the main contributions to small-scale interface roughness.[12] In heterostructures built from A and B semiconductors, the A/B and B/A interfaces have been shown to be nonequivalent as far as interface roughness is concerned.[2,4,6,12]

The existence of interface islands is considered to be the main cause of exciton localization in QW's and SL's. Although localized excitons probe interface defects on a length scale much larger than the atomic scale, in this paper we show that the study of excitonic photoluminescence with regard to the polarization properties of emitted light brings useful information on the interface structure, the dissymetry of adjacent interfaces, and the localization process. Type-II QW's and SL's are very appropriate for this study, since electrons and holes are confined in adjacent layers, and their envelope functions overlap at interfaces.

The present work is based on results obtained from previous studies of the fine structure of excitonic states in type-II pseudodirect SL's by PL and optically detected magnetic resonance (ODMR).[13–16] The electron–heavy-hole pair state is built from a Γ-hole state and an X_z-electron state. The X_z-electron state in AlAs is coupled to the Γ-electron state in GaAs, making the optical transition weakly allowed.[17] The electron–heavy-hole pair state is fourfold degenerate. This degeneracy is lifted by the electron-hole short-range exchange interaction leading to a Γ_5 exciton radiative doublet and to Γ_1 and Γ_2 exciton nonradiative states[18] separated from the Γ_5 state by the exchange energy ε (Fig. 1). Recently it was established experimentally that the degeneracy of the Γ_5 excitonic doublet is lifted in type-II GaAs/AlAs superlattices.[13,14] The experimental results are well explained by the theory developed by Aleiner and Ivchenko[19] if one assumes that excitonic states are localized at interfaces. Very likely both the electron and hole are localized by layer thickness fluctuations, and the electron recombines with a hole localized either in the right or left GaAs layer, i.e., at the inverted interface (GaAs on AlAs) or at the direct interface (AlAs on GaAs), respectively. Aleiner and Ivchenko showed that, for a single A/B or B/A type-II quantum well, the exciton radiative doublet is split into [110] and [110] dipole-

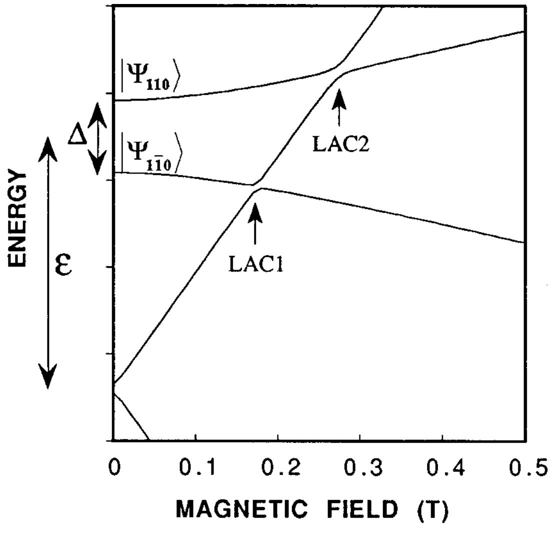

FIG. 1. Heavy exciton energy levels of a type-II SL in a longitudinal magnetic field. $|\Psi_{110}\rangle$ and $|\Psi_{1\bar{1}0}\rangle$ are the exciton radiative levels. Only the case with $\Delta > 0$ is represented. The level anticrossings LAC1 and LAC2 between the upper nonradiative state and the two radiative ones are indicated by arrows.

active states owing to the combined effect of exchange interaction and mixing of heavy- and light-hole states at the interface. These radiative states are denoted hereafter as $|\Psi_{110}\rangle$ and $|\Psi_{1\bar{1}0}\rangle$, respectively (Fig. 1). Δ is their splitting energy, in the range of a few μeV. The absolute value of Δ is experimentally obtained from the period of quantum beats observed during the decay of excitonic PL when the transition is resonantly excited with [100]-polarized light.[14,20] It was predicted in Ref. 19 that the sign of Δ (or in other words the ordering of $|\Psi_{110}\rangle$ and $|\Psi_{1\bar{1}0}\rangle$ states) is opposite for A/B and B/A interfaces. This sign is then characteristic of the type of interface (direct or inverted) at which excitons are localized. Experimentally, it was shown from quantum beats studies that excitons are mainly localized at one kind of interface, but neither the sign of Δ nor the nature of this interface could be determined.[14,19]

To obtain experimentally the sign of the splitting energy Δ and its correspondence with the nature of the interface, we performed magneto-optical studies of type-II pseudodirect SL's of various compositions. Let us consider excitons localized at interfaces of one type (with $\Delta > 0$, for instance). In a longitudinal magnetic field directed along the growth direction (z axis), the Zeeman splitting of the excitonic states leads to the level scheme of Fig. 1. For two values of the field the upper nonradiative level crosses the two radiative levels. As a matter of fact, state mixing by any small in-plane component of the field or by hyperfine interaction results in level anticrossing (LAC).[21] Let us consider specifically the experimental situation where the four excitonic sublevels are populated after relaxation of electron-hole pairs created by band-to-band excitation. With increasing magnetic field, at LAC fields, the oscillator strength becomes equally shared between the exciton states. At these particular field values, owing to the larger population of the nonradiative state the intensity of emitted light increases.[21] Let us consider now the polarization of emitted light. With increasing field the polarization of light emitted by the radiative states changes from ([110], [$1\bar{1}0$]) linear polarization to (σ_+,σ_-) circular polarization. Since in the range of field considered here the splitting of the radiative states is much smaller than the PL linewidth, the detected light originates from both radiative states. Therefore the degree of polarization of PL, either linear or circular, is nearly equal to zero. However, at LAC fields, the additional emitted light is elliptically polarized with the long axis of the ellipse along the direction of polarization of the lowest (upper) radiative level at the first (second) LAC field. Hence by measuring the degree of linear polarization of PL as a function of the magnetic field we are able to determine the ordering of the zero-field excitonic radiative states $|\Psi_{110}\rangle$ and $|\Psi_{1\bar{1}0}\rangle$. The correspondence between the sign of Δ and the interface at which excitons are localized is obtained from the study of type-II samples in which excitonic recombination is forced at one kind of interface by the design of the structure.

This paper is organized as follows. In Sec. II we describe the samples and the experimental setup. In Sec. III we report on level anticrossing results. We relate them to the growth conditions of the samples, and we discuss exciton localization with respect to interface roughness. We also discuss PL polarization at zero field. Finally, in Sec. IV we summarize the results and conclude.

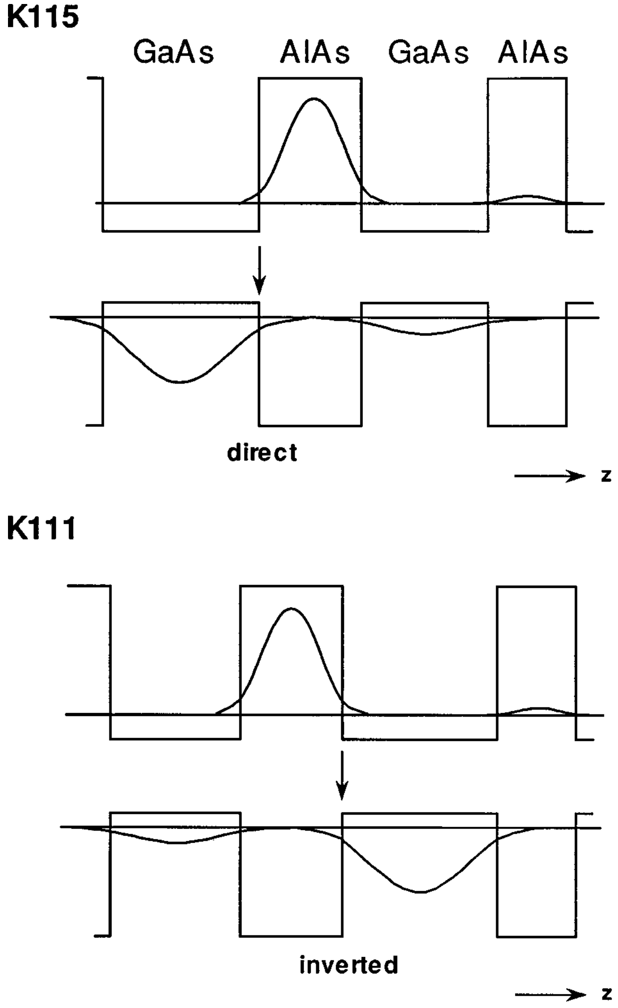

FIG. 2. Schematic representation of the band structure and envelope functions for electrons and holes in samples $K115$ and $K111$.

II. SAMPLE CHARACTERIZATION AND EXPERIMENTAL SETUP

The samples were grown by molecular-beam epitaxy. Samples $K111$ and $K115$, designed to reveal the correspondence between the sign of Δ and the interface of localization, are composed of 336 periods of a SL grown on top of a n^+-doped GaAs substrate and n^+-doped GaAs buffer layer. They are capped with 100 nm of n^+-GaAs. The nominal composition of one SL period is (GaAs 15 Å/AlAs 12 Å/GaAs 18 Å/AlAs 9 Å) for $K111$, and (GaAs 18 Å/AlAs 12 Å/GaAs 15 Å/AlAs 9 Å) for $K115$. Thus the sequence of layers is identical for the two samples except for the inversion of the growth direction. The band structure is schematically shown in Fig. 2. In both samples electrons and holes are mainly confined in the 12-Å AlAs layer and in the 18-Å GaAs layer, respectively. The excitonic recombination is then expected to occur at the same energy but at the (GaAs on AlAs) inverted interface for $K111$ and at the (AlAs on GaAs) direct interface for $K115$. As a matter of fact x-ray diffraction, Raman, and PL results show that the composition of each sample is slightly different from the nominal one. However this has no incidence on the validity of the LAC results. From x-ray diffraction one finds that the total AlAs

TABLE I. Sample composition and growth parameters. The column on the far right shows the result obtained from magneto-optical studies, i.e., the type of interface at which exciton recombination occurs (see text).

Sample	Composition GaAs(Å)/AlAs(Å)	Doping of the substrate/buffer	Growth interruption	Substrate temperature °C	Growth rate (Å)/s GaAs/AlAs	Interface of recombination
E219	18/12	*n*-type/*n*-type	no	640	2/1	direct
D207	28/20	*n*-type/*n*-type	no	640	2/1	direct
E913	20/11	no/no	no	640	2	direct
B223	17*A*/26	*n*-type/no	no	650	1.35/1.75	direct
A5L23 SL1	25/22	no/*n*-type	yes	580	2/1	direct
A5L23 SL2	20/15	no/*n*-type	no	580	2/1	inverted
B321	31/23.6	*n*-type/no	no	600	0.5	inverted
B209	24/13.6	*n*-type/no	no	600	0.5	inverted

thickness in one period is equal to 20.4 Å (*K*111) and 20.6 Å (*K*115), very close to the nominal one (21 Å). The total GaAs thickness in one period is smaller for *K*115 (28.8 Å) than for *K*111 (31.3 Å). Both values are smaller than the nominal one (33 Å). Raman spectra show that the AlAs LO modes are identical in the two samples whereas the GaAs LO mode frequencies are consistent with thinner layers in *K*115 than in *K*111. In accordance with these results, the PL spectrum is at slightly higher energy for *K*115 than for *K*111.

A set of nominally symmetrical SL's, with a two-layer period, was also investigated in order to determine the influence of growth conditions on the exciton localization at one interface or the other. The samples composition and growth conditions are given in Table I. For each sample the [110] and [110] directions were either revealed by surface reconstruction during growth or checked afterwards by examining the sample surface with an optical microscope with magnification *X*400, looking for the so-called ''oval'' defects whose orientation with respect to the [110] and [110] directions is well known. We have examined the oval defects of *B* type according to Fig. 2 of Ref. 22.

The PL was excited with the 514.5-nm line of an argon laser. PL light was dispersed in a monochromator and detected by a photomultiplier followed by a lock-in amplifier. The polarization of PL light detected at the peak of the excitonic PL line was analyzed by using a photoelastic modulator. The amplitude of modulation at frequency *f* applied along the *x* axis of the modulator is adjusted in order to obtain a maximum dephasing of π between the *x* and *y* axes of the modulator. The modulator is followed by a polarizer aligned along the [110] axis of the sample and at 45° of the *x* axis of the modulator. We obtain the difference of PL intensities $[I_{[1\bar{1}0]}-I_{[110]}]$ at frequency $2f$. The PL intensity polarized along the [110] direction is obtained by inserting a polarizer parallel to the [110] direction in between the sample and the modulator. The experimental setup used to observe PL quantum beats in order to obtain the splitting energy Δ has been described elsewhere.[14] The sample was mounted on a sample holder placed in the core of a small superconducting coil immersed in a standard optical cryostat at pumped liquid-helium temperature.

III. RESULTS AND DISCUSSION

A. LAC results for samples *K*111 and *K*115

The LAC signal is shown in Fig. 3(a) for samples *K*111 and *K*115. The difference of PL intensities $I_{[1\bar{1}0]}$ and $I_{[110]}$ with polarizations parallel to the [110] and [110] directions, respectively, is plotted as a function of the magnetic field. For sample *K*115, where exciton recombination occurs at the direct interface, one observes a decrease followed by an increase of the signal. We conclude that the lowest excitonic level has [110] symmetry. The opposite conclusion is reached for sample *K*111: the lowest level has [110] symmetry at the inverted interface.

The field values at the anticrossings as well as the difference between them are larger for sample *K*115 than for

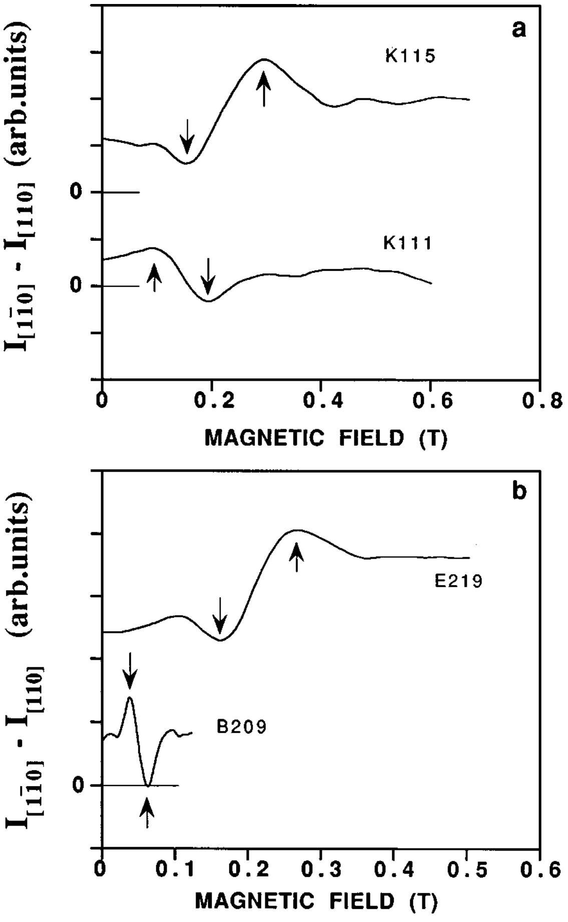

FIG. 3. Level anticrossing signals for samples *K*115 and *K*111 (a) and for two-layer superlattices: samples *E*219 and *B*209 (b). In (a), the vertical scale has been shifted for sample *K*115 for the sake of clarity.

sample $K111$. This is consistent with a larger value of Δ and a larger exchange energy ε for sample $K115$ than for sample $K111$. The value of Δ is obtained from the period of the quantum beats observed during the PL decay. It was found equal to 11.8 ± 0.6 and 9.4 ± 0.4 μeV for samples $K115$ and $K111$, respectively. The value of ε was obtained from ODMR measurements.[16] It was found equal to 23.8 and 19 μeV for samples $K115$ and $K111$, respectively. These results are in agreement with the fact that GaAs layers are thinner in $K115$ than in $K111$. As a consequence, the overlap between the electron and the hole envelope function is larger in $K115$ and both the exchange energy ε and the splitting energy Δ are larger.

To our knowledge the one-to-one correspondence between the ordering of $|\Psi_{110}\rangle$ and $|\Psi_{\overline{110}}\rangle$ states and the nature of the interface has not yet been established theoretically. Calculation of the heavy-hole–light-hole mixing at interfaces within a tight-binding approach should give the correct ordering of excitonic levels provided that the value and sign of the tight-binding parameters for GaAs and AlAs are known with sufficient accuracy. Nevertheless, simple qualitative considerations can help to understand the experimental results. In a GaAs layer the projections of the arsenic-gallium bonds on the (001) plane lie along the [110] direction at the indirect interface, whereas they lie along the [110] direction at the direct interface. To some extent, the hole Bloch function retains the p character of the arsenic-gallium bond. Therefore, at the inverted interface it has a p_x component with $x \| [110]$ and at the direct interface a p_y component with $y \| [110]$. Since the optical selection rules for the excitonic transition are governed by the symmetry of the hole Bloch function, the experimental results are qualitatively understood, i.e., the lowest excitonic level is dipole active along the [110] ([110]) direction at the inverted (direct) interface.

B. Results in nominally symmetrical SL's

We have investigated LAC signals in a set of samples grown in various conditions. Typical LAC curves are displayed in Fig. 3(b). Each studied sample exhibits a LAC signal with linear polarization. This is a clear indication that, for a given sample, excitons are mainly localized at one kind of interface. If they were localized at random at one or the other interface, contributions of the two classes of excitons with $\Delta > 0$ and $\Delta < 0$ would not lead to any LAC signal with linear polarization.

The results are summarized in Table I. Samples are classified into two categories according to the results of LAC measurements: samples with recombination at the direct interface or samples with recombination at the inverted interface. The composition of the SL, i.e., the thickness of GaAs and AlAs layers, does not seem to play a role in this classification since in samples with close compositions (like $E913$ and $A5L23$-SL2) excitons do not recombine at the same interface. The possible electric field due to the doping of the substrate and/or the buffer does not play a role either, as can be inferred from Table I. Conversely, growth temperature and growth interruption (GI) at interfaces play a key role. The influence of the latter is especially obvious for sample $A5L23$ in which two superlattices SL1 and SL2 have been grown at the same temperature, but one of them with GI at interfaces and the other one without GI. Excitons in SL1 with GI recombine at the direct interface, and excitons in SL2 without GI recombine at the inverted interface. It can be deduced from the results of Table I that excitons recombine at the direct interface either when the sample was grown with GI or with a high substrate temperature (640 °C).

Recombination at the direct or inverted interface is determined by the localization of excitons or electron-hole pairs. It is clear that, in type-II SL's, most of the PL signal arises from the recombination of excitons and not of distant electron-hole pairs since the exchange interaction measured in ODMR experiments is typical of type-II excitons.[13,15]

Exciton localization at the direct or at the inverted interface cannot be explained by the asymmetry of interface profiles resulting from gallium segregation during growth.[12] We have theoretically considered a superlattice in which an enlarged GaAs layer and an enlarged AlAs layer, adjacent to it, are included. The enlarged GaAs layer is located prior to or after the enlarged AlAs layer in order to simulate exciton localization at the normal or inverted interface, respectively. Taking segregation into account, we calculated the variation of confinement energies of the electron and hole, and we estimated the difference of exciton binding energy E_b in the two configurations. For the composition of our samples, the electron confinement energy does not change with the configuration but the hole confinement energy is smaller by 0.5–1.5 meV for localization at the direct interface. On the other hand, the exciton binding energy E_b is larger by at most 4% for localization at the inverted interface. In order to obtain localization of the exciton at the inverted interface, the energy variation ΔE_b should counterbalance the variation of the hole confinement energy. This is hardly realized in our samples. Segregation could thus explain exciton localization at the direct interface but not at the inverted one. Moreover, the calculation shows almost no dependence of the hole confinement energy and the exciton binding energy on the variation of the potential profile with growth temperature. This is not in agreement with the experimental results which show that the interface at which excitons are localized depend very much on the growth temperature. Therefore, we conclude that our results cannot be explained by the effect of segregation.

We believe that exciton localization at the direct or inverted interface is determined by the in-plane extension of interface defects. As the exciton binding energy in GaAs/AlAs type-II superlattices is large inspite of the small overlap of envelope functions,[23,24] the two carriers cannot be far apart. They have to be localized at the same in-plane position, on both sides of an interface. Which region localizes the exciton is determined by the decrease of confinement energy for both carriers. Since the X_z electron and the heavy-hole effective masses are of comparable magnitudes, the localizing potential for the exciton center of mass is not the localizing potential for one or the other carrier, but an average of the localizing potentials for both carriers over the electron and hole in-plane relative motion. As a consequence, only defects with sizes in the range of or larger than the exciton Bohr radius can localize excitons. Defects with size and separation much smaller than the Bohr radius result in a scattering potential.

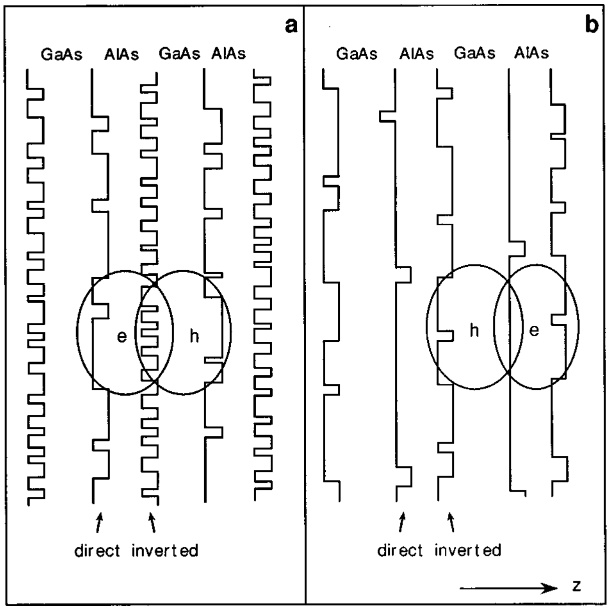

FIG. 4. Schematic representation of exciton localization by interface roughness for different growth conditions. (a) Low substrate temperature and no growth interruption at interfaces. (b) High substrate temperature or growth interruption at interfaces. Residual atomic-scale roughness has not been represented.

In Fig. 4 we schematically represent the exciton localization process. We assume interface islands of 1-ML thickness, and no correlation of the position of defects from one layer to the next. The length scale of interface islands is known to be larger at the direct interface than at the inverted one,[2,4,6] owing to the larger atomic diffusion coefficient of gallium with respect to that of aluminium. For a sample grown at low temperature and without GI, the inverted interface exhibits defects at a scale too small to be effective in localizing carriers. They are then localized by defects at the direct interface [Fig. 4(a)]. Since an increase of the effective layer thickness in one layer corresponds to a decrease of the effective thickness in the adjacent layer next to the direct interface, the electron and the hole very likely do not localize on both sides of the direct interface. We can conclude, in agreement with experiments, that in these samples excitons recombine at the inverted interface. With higher growth temperature or introduction of GI at interfaces, the roughness of the direct interface decreases, and the length scale of interface islands at the inverted interface increases [Fig. 4(b)]. Defects at the inverted interface become effective for the localization of carriers. For the same reason as above, the electron and hole do not localize on both sides of the inverted interface. The excitonic recombination then occurs at the direct interface, as depicted in Fig. 4(b). This is in agreement with the experimental results. We conclude that the LAC measurement indeed gives a signature of the quality of interfaces.

C. Zero-field degree of polarization

Even without applied magnetic field, the study of the degree of polarization of photoluminescence brings information on carrier localization and the geometry of interface defects. For each sample studied we have found at zero magnetic field a small degree of linear polarization along the $[1\bar{1}0]$ direction ($[I_{[1\bar{1}0]}-I_{[110]}]>0$). This is clearly seen in Fig. 3. The degree of polarization is 1–3% at the peak of the zero-phonon PL line. It varies inside the PL line and increases toward the low-energy side, as shown in Fig. 5.

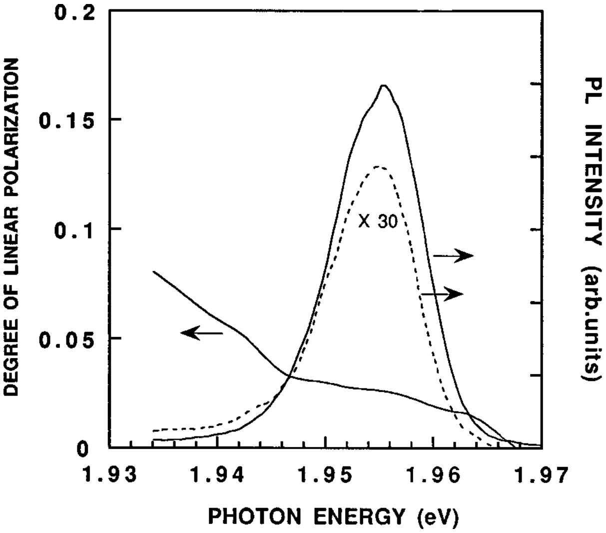

FIG. 5. Photoluminescence and degree of linear polarization along the [110] direction for sample K115 under band-to-band excitation and at zero magnetic field. Solid PL line: sum of PL intensities $I_{[1\bar{1}0]}$ and $I_{[110]}$ with polarization along [110] and [110]; dashed PL line: difference of PL intensities $I_{[1\bar{1}0]}$ and $I_{[110]}$.

ODMR results obtained on some of the samples listed in Table I bring complementary information on the electronic states.[15] A decrease of the exchange energy is observed in the low-energy part of the PL line, where the degree of polarization is larger. The spectrum of the free-electron ODMR signal is also shifted to the low-energy part of the PL line with respect to the spectrum of the exciton signal. These results show that radiative recombination in the low-energy tail of the zero-phonon PL line involves (quasi) free electrons. We tentatively attribute the [110]-polarized weak PL signal to the recombination of distant electron-hole pairs, with the hole localized in anisotropic defects elongated along the [110] direction. The anisotropic localizing potential for the hole mixes the heavy- and light-hole states. As a result of the anisotropy PL exhibits a nonzero degree of polarization along the [110] direction.[25]

Another possible explanation that cannot be completely ruled out is the effect of the electron-hole long-range exchange Hamiltonian on excitons localized by anisotropic defects elongated along the [110] direction. For free excitons the long-range exchange interaction leads to the well-known longitudinal-transverse splitting. For localized excitons, the degeneracy of the two exciton states which are dipole allowed for polarization along the [110] and [110] directions, is lifted and the radiative probability $W_{[1\bar{1}0]}$ for the [110] level is larger than the radiative probability $W_{[110]}$ for the [110] level.[26] From numerical estimations of $W_{[1\bar{1}0]}$ and $W_{[110]}$,[27] one expects a degree of linear polarization of PL of a few percent along [110], which is also consistent with our experimental results. Whatever the exact interpretation, the experimental results are in agreement with the observation of interface defects elongated along [110] by scanning tunneling microscopy.[28,29]

IV. CONCLUSION

We have demonstrated that the fine structure and polarization properties of the excitonic PL in type-II pseudodirect superlattices can be used to obtain information on the localization process of excitons and on interface characteristics. The wave functions of localized excitons do not extend over many interfaces. In every sample studied, excitons recombine mainly at one kind of interface, either the direct one or the inverted one depending on the growth conditions. We have demonstrated, on SL's where the interface of recombination is determined by the sample design, that, for excitons recombining at the direct (inverted) interface, the lowest exciton state is dipole active for light polarization along the [110] ([110]) direction.

We have studied the influence of growth conditions on the quality of interfaces. In our series of samples, excitons recombine at the direct interface either when the sample is grown with growth interruption at interfaces or with high substrate temperature (640 °C). They recombine at the inverted interface when the sample is grown at lower temperature or without growth interruption. In both cases, excitons recombine at the interface with defects at a length scale such that they cannot localize the exciton, either because the interface is flat over sufficient distance or because the size and separation of islands are too small. We believe that the method used in this work to demonstrate the dissymetry of normal and inverted interfaces with regards to exciton localization and recombination could be readily applied to other type-II heterostructures, provided that the heavy exciton level scheme in a magnetic field is similar.

Additional information on carrier localization was obtained from the small degree of linear polarization along the [110] direction observed in the low-energy tail of the PL line for every sample at zero field. This weak [110]-polarized PL signal is attributed to the recombination of distant electron-hole pairs with the hole localized in anisotropic defects elongated along the [110] direction.

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