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Effects of Be acceptors on the spin polarization of carriers in p-i-n resonant tunneling diodes

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In this paper, we have investigated the effect of Be acceptors on the electroluminescence and the spin polarization in GaAs/AlAs p-i-n resonant tunneling diodes. The quantum well emission comprise two main lines separated by ∼20 meV attributed to excitonic and Be-related transitions, which intensities show remarkably abrupt variations at critical voltages, particularly at the electron resonant peak where it shows a high-frequency bistability. The circular-polarization degree of the quantum-well electroluminescence also shows strong and abrupt variations at the critical bias voltages and it attains relatively large values (of ∼−75% at 15 T). These effects may be explored to design novel devices for spintronic applications such as a high-frequency spin-oscillators. *© 2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4891996]

I. INTRODUCTION

Voltage control of the spin polarization of carriers is an important issue for the development of novel spintronic devices.[1,2] Particularly, it was shown that the spin-polarization of carriers in Resonant Tunneling Diodes (RTDs) can be voltage-selected, which makes these devices very attractive for spintronic applications.[3–15] The successful operation of semi-magnetic RTDs as a voltage-controlled electron-spin-polarized filter was first reported for structures based in II–VI semiconductor alloys with a magnetic ion.[4] Spin effects in III–V non-magnetic RTDs under high magnetic was previously investigated.[5–15] It has been demonstrated that both the excitonic spin-splitting energy and the polarization degree of the quantum well (QW) emission from p-i-p non-magnetic RTDs under high magnetic fields present strong oscillations near resonant peak voltages. In addition, the spin-splitting and circular polarization degree did not present a clear correlation, which implies that the spin polarization of carriers in the QW cannot be explained by a simple thermal occupation of the QW confined levels. Therefore, other effects such as spin-conservation along the resonant tunneling through spin-split levels and the spin-polarization of the carriers accumulated at the contact layers should also be considered in order to explain the spin-polarized emission from these devices.

In this paper, we have investigated the effect of Be acceptors in the QW on the optical and spin properties of a GaAs/AlAs p-i-n resonant tunneling diode. We have performed polarization-resolved magneto-electroluminescence (EL) measurements under magnetic fields up to 15 T. We observed two main QW EL peaks separated by ∼20 meV, which are attributed to the excitonic recombination between fundamental confined states in the QW (E1 – HH1 recombination) and to the recombination involving holes bound to acceptors in the QW (E1-A_0 recombination). We also observed remarkably abrupt changes on the relative intensities of these two emission lines at critical voltages that correspond to the onset of electron resonant tunneling and to the E1 resonant peak. This effect is accompanied by similar abrupt changes of the QW polarization degree near the E1 resonant voltage peak. Particularly, the device shows evidence of a spin polarization bistability at this critical voltage which may be interesting for new spintronics devices.

II. EXPERIMENT DETAILS

Our RTD was grown by Molecular Beam Epitaxy on a (001) n+ GaAs substrate. The device consists of the following layers: 3 μm p-GaAs (2×10^{18} cm^{-3}) bottom contact, 100 nm p-GaAs (1×10^{18}cm^{-3}), 100 nm p-GaAs (5×10^{17}cm^{-3}), 51 Å undoped GaAs spacer, 51 Å undoped AlAs barrier, 59 Å undoped GaAs QW, 51 Å undoped AlAs barrier, 76 Å undoped GaAs spacer, 348 Å n-GaAs (10^{16}cm^{-3}), 508 Å n-GaAs ($2 \times$ cm^{-3}), 1 μm n-Al$_{0.33}$Ga$_{0.67}$As (1.33×10^{18} cm^{-3}) optical window, and 254 Å n-GaAs (2×10^{18} cm^{-3}) top contact. P-type and n-type layers were achieved using Be impurities and Si, respectively. It is worth to point out that the thickness of the spacer layers are only ∼5 nm. Therefore, besides background impurities, acceptors can diffuse into the QW during growth from the doped layers. This effect is expected to be stronger for Be diffusion from the p-doped layers as they are grown first, i.e., before the double-barrier structure. Circular mesas with 400 μm diameters were processed with annular AuGe contacts to allow optical measurements. Eletroluminescence measurements were performed at low temperatures under magnetic fields up to 15 T using a Si CCD system coupled to a 0.5 m Andor spectrometer.

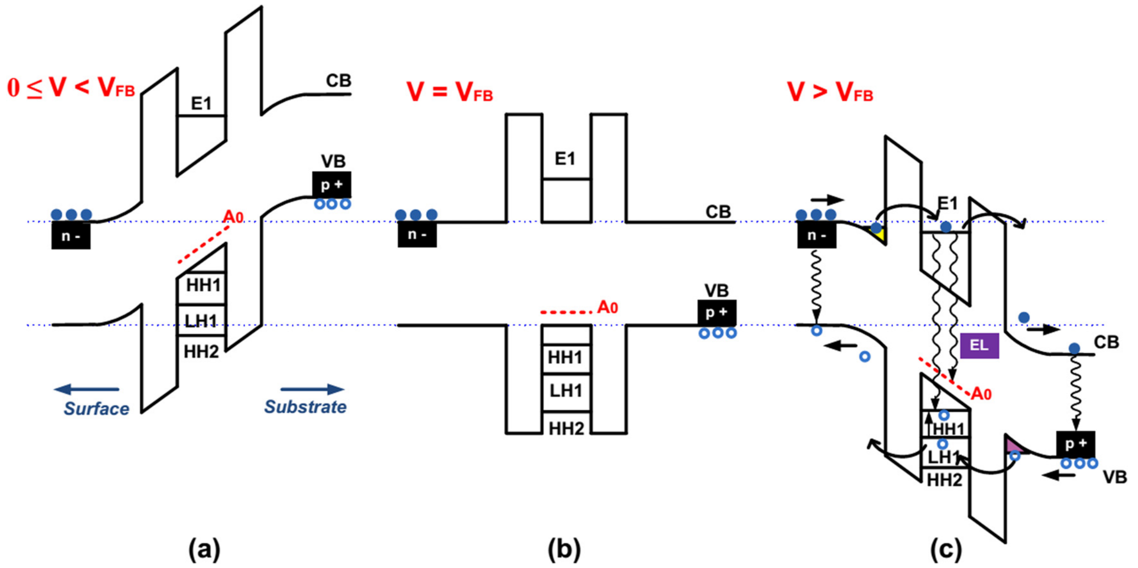

FIG. 1. Schematic band diagram of the device including confined states (E1, HH1, LH1, and HH2) and a shallow level A0 due to the diffusion of Be impurity in the QW for (a) voltages lower than flat band condition (b) at flat band condition and (c) for voltages higher than flat band condition.

Right- (σ^+) and left- (σ^-) circularly polarized emissions were selected by using appropriate optical components.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 1 shows a schematic band diagram of our structure for: (a) voltages lower than the flat band condition (labeled V_{FB}) ($V < V_{FB}$), (b) for the flat band condition ($V = V_{FB}$); and (c) under a forward bias larger than V_{FB} ($V > V_{FB}$). Under low applied voltages ($V \leq V_{FB}$), a negligible current flows through the device, which corresponds to a typical current in a reverse biased diode. However, as the voltage is increased attaining a forward bias condition ($V > V_{FB}$), quasi-two-dimensional (2D) gases of electrons and holes form at the spacer layers between the barriers and the doped layers for the n-type and p-type sides, respectively. Under this condition, carriers from the 2D gases, including electrons (E), light- (LH), and heavy- (HH) holes can resonantly tunnel through the QW. At this condition, a current thus flows through the structure and an EL signal can be observed from both the GaAs contact layers and the QW. As will be discussed later, depending on the applied bias voltage, the QW emission is dominated by either an free excitonic transition (E1–HH1 recombination) or to a recombination involving Be acceptors (E1-A_0 recombination), as illustrated in Figure 1.

Figure 2 shows a typical characteristics I(V) curve of the p-i-n RTD at low temperatures (5 K) where pronounced electron- and hole-related resonances are evidenced. Particularly, three clear peaks are observed, which we attributed to the resonant tunneling through the LH1, E1, and HH2 states of the QW. In addition, a weak HH1 shoulder is observed at lower bias solely in the differential conductance of the I(V) characteristic curve (inset). The flat band condition is obtained when the p-i-n diode is forward biased with ~1.51 V.

Figure 3(a) presents a color-coded map of the QW EL intensity as function of applied voltage under B = 0 T. As mentioned before, under applied voltage, carriers from the contact layers can tunnel through the QW and an EL signal is then observed from this region. For an applied bias under B = 0 T, the $\sigma+$ and the $\sigma-$ circularly polarized QW EL intensities are identical. Therefore, the polarization degree is zero for B = 0 T. The voltage dependence of the total QW integrated EL intensity including both emission bands is shown in Fig. 3(b). The total QW emission intensity presents a good correlation to the I(V) characteristics curve. If we consider a simple theoretical model, the QW EL intensity should be proportional to the product of the densities of electrons and holes inside the QW. Therefore, the abrupt EL rising at ~1.66 V approximately coincides with the onset of resonant tunneling of electrons into the QW (E1), when a significant number of holes must already be tunneling into the LH1 level. Furthermore, at the LH1 and E1 resonant peaks, the EL intensity decreases abruptly. For high biases (>1.85 V), the correspondence weakens out as the probability of carriers tunnelling out of the QW through the double barriers increases, which gives rise to a current but decreases the density of carriers accumulated in the QW.

The EL image (Fig. 3(a)) and the selected spectra presented in Fig. 3(c) clearly show that the QW emission comprises two main bands separated by ~20 meV. The relative intensity of these two bands present very abrupt variations

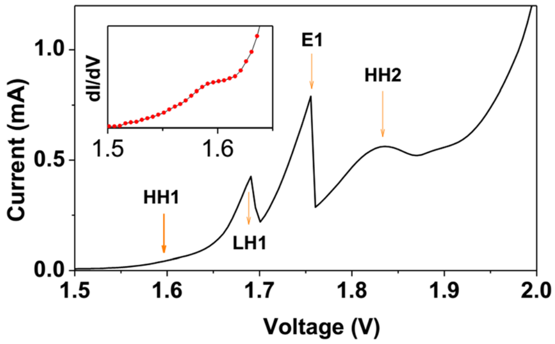

FIG. 2. (a) I(V) characteristics curve at 5 K. The insert shows the differential conductance dI/dV.

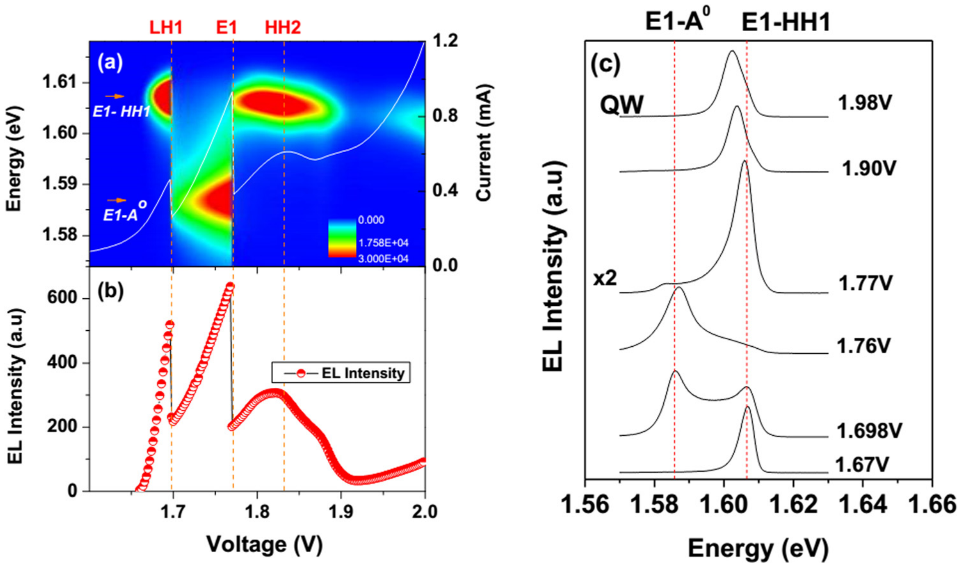

FIG. 3. (a) Color-coded map of QW EL intensity as a function of applied voltage; (b) Voltage dependence of the total QW EL integrated intensity; (c) Typical electroluminescence spectra of QW at 2 K for different applied voltages.

that also correlate with the I(V) curve. The lower energy band dominates the emission only for biases between the LH1 and the E1 resonant peaks, where the tunneling of electrons into the QW must overshadow the tunneling of holes. For all other biases, the EL emission shows only the higher energy band.

The high energy band is consistent with the calculated energy of the excitonic recombination involving carriers from the fundamental confined states of the QW (E1-HH1). Therefore, the lower-energy emission must involve some bound state in the QW. An analogous transition was previously observed using similar RTD devices.[16] This previous work[16] conclusively demonstrated that the lower energy band is related to transitions involving Be acceptors that can diffuse from the p-doped contact into the QW during the growth process of the devices. Particularly, they have shown that increasing the spacer layer thickness between the Be doped region and the tunnel barriers greatly reduces the intensity of the lower energy emission band. In addition, they have shown that the introduction of an intentional Be delta doping in the QW of a p-i-n RTD strongly enhances the relative intensity of this emission. Furthermore, the energy difference between these EL emission lines observed in our case, as well as in the previous work,[16] is of ~20 meV. We can consider the simple expression for the binding energy of a shallow acceptor in a QW

$$E_{A0} = E_{E1-HH1} - E_{E1-A0} + E_X,$$

where $E_{E1\text{-}HH1}$ is the energy of the fundamental excitonic QW transition, $E_{E1\text{-}A0}$ is the energy of the transition involving E1 electrons and holes bound to this shallow acceptor, and E_X is the excitonic binding energy. Therefore, using the experimental result ($E_{E1\text{-}HH1}$ - $E_{E1\text{-}A0}$) ~ 20 meV and the excitonic binding energy (~17 meV) for a similar GaAs/AlAs QW,[17,18,29,30] we obtain $E_{A0} = 37$ meV, which is in agreement with previously theoretical/experimental result $E_{A0} \sim 38$ meV, reported for Be acceptors binding in similar GaAs/AlAs QWs.[16,26–28] However, there is an uncertainty on the value of E_{Be} because the binding energy is actually strongly dependent on the exact position of the acceptors in the QW. Therefore, we attribute the observed lower energy EL band to the recombination of free electrons and holes bound to neutral Be acceptors in the QW, which we label as the E1-A0 recombination.

Since the advent of semiconductor QWs, it has been noticed that the observation of a shallow acceptor-related transition from a QW has a relatively low probability as compared to a bulk material with similar impurity concentrations. This distinction has been attributed to the restriction of the QW confined carriers to a more limited volume as compared to bulk, especially for QWs with interface roughness, when the carriers remain at trapped at interfaces localized states at low temperatures. In bulk, carriers probe a larger volume and therefore have a larger probability of being eventually captured by a lower energy state related to a residual impurity resulting in a more efficient recombination through impurity-related states as compared to QWs.[19–22] However, the probability of impurity related transitions in a QW can be significantly increased by reducing the carrier localization, for instance by a small increase of the temperature or by increasing the density of the carriers, as those effects give rise to an increased occupation of less localized QW states.[16] Therefore, the significant variations of carriers density in the QW with small variations of the bias voltage applied to a RTD structure may induce abrupt variations of the intensity of QW impurity related transitions. In our case, we believe that the abrupt appearance/disappearance of the acceptor-related transition can be associated to the sudden increase/ decrease of the density of injected electrons in the QW, resulting in the occupation/emptying of delocalized QW states where the electrons are more efficiently captured by residual Be impurities in the well.

The evolution of the EL emission is particularly abrupt around the E1 resonant peak, when the spectrum is immediately transformed from a single low energy band (~1.585 eV) to one clearly dominated by the higher energy band (~1.605 eV). In fact, the density of electrons in the QW must decrease very abruptly when the applied voltage makes the fundamental state of the two dimensional electron gas (2DEG) overpasses the E1 level, usually known as the negative-differential resistance condition. Moreover, a high-frequency bistability effect is commonly observed in such condition resulting in abrupt variations of the current and in a hysteresis-like effect.[23,24] In our case, the bistability is accompanied by a remarkable variation of the EL emission with a substantial energy shift of ~20 meV. This effect can be observed by recording the EL emission as a function of time using a streak camera detector (Hamamatsu C4334). Figure 4 shows a single image shot of the QW EL emission during a 10 ms time interval while the RTD structure is submitted to a constant bias voltage (1.7585 V) corresponding to the negative-differential region (NDR) associated to the E1 resonant peak. We also present two typical spectra obtained by integrating the EL emission over two distinct 0.5 s time intervals and the integrated intensity for 2 nm intervals at the peak of each band *versus* time. The spectra always show the two QW emission bands discussed above. However, the intensity of the low energy band presents a spontaneous abrupt intensity variation at a given instant of the measurement, which is clearly revealed by the intensity versus time plot. Various single shot measurements were taken and several of them present one or more similar random transitions where the intensities of the two bands present sudden intensity variations. The observed EL telegraphic-like noise is a remarkable reflection of the bistability of the RTD structure that results in a strong abrupt variation of the density of electrons in the QW.

Figures 5(a) and 5(b) present color-coded maps of the circularly polarized ($\sigma+$ and $\sigma-$) QW EL emission as a function of the applied bias and the I(V) characteristic curve at 15 T. Besides the resonant peaks associated to electron and hole resonances discussed above, the I(V) curve show additional peaks after the E1 peak associated to the well-known scattering-assisted resonant magneto-tunneling effect.[25] The EL emission maintains the same as those obtained without magnetic field, with the low energy QW band dominating the spectra solely while the electron injection into the QW is resonant. We have used the same intensity color scale for both, $\sigma+$ and σ^- images, making evident that the EL emission is σ^- polarized. We also present the total integrated QW EL emission intensities as a function of the bias voltage (Figure 5(c)), which clearly correlates with the I(V) curve and reinforces the fact that the emission is strongly σ-polarized for voltages smaller than the E1 resonance.

Figure 5(d) presents the voltage dependence of the experimental degree of circularly polarization (DCP) calculated by

$$\mathrm{DCP} = (\mathrm{I}^{\sigma+} - \mathrm{I}^{\sigma-})/(\mathrm{I}^{\sigma+} + \mathrm{I}^{\sigma-}), \tag{1}$$

where $\mathrm{I}^{\sigma+}$ and $\mathrm{I}^{\sigma-}$ are the integrated intensities of the $\sigma+$ and σ^- circularly polarized QW EL emission (shown in Figures 5(a) and 5(b)). The DCP was calculated for all measured bias voltages considering the total integrated EL emission including both energy bands and gives information about the spin polarization of the carriers in the QW. The experimental DCP should represent the averaged spin polarization of the distributions of electrons and holes in the QW, considering the established selection rules.[2] Furthermore, we have also estimated the separated polarization degree for each emission band by fitting the EL spectra with Gaussian bands for some bias voltages. This estimation involves, however, a relatively large error as the bands are not well resolved and usually the EL emission is strongly dominated by one of the bands, so that the other band appears solely as a small shoulder of the main emission. In general, the

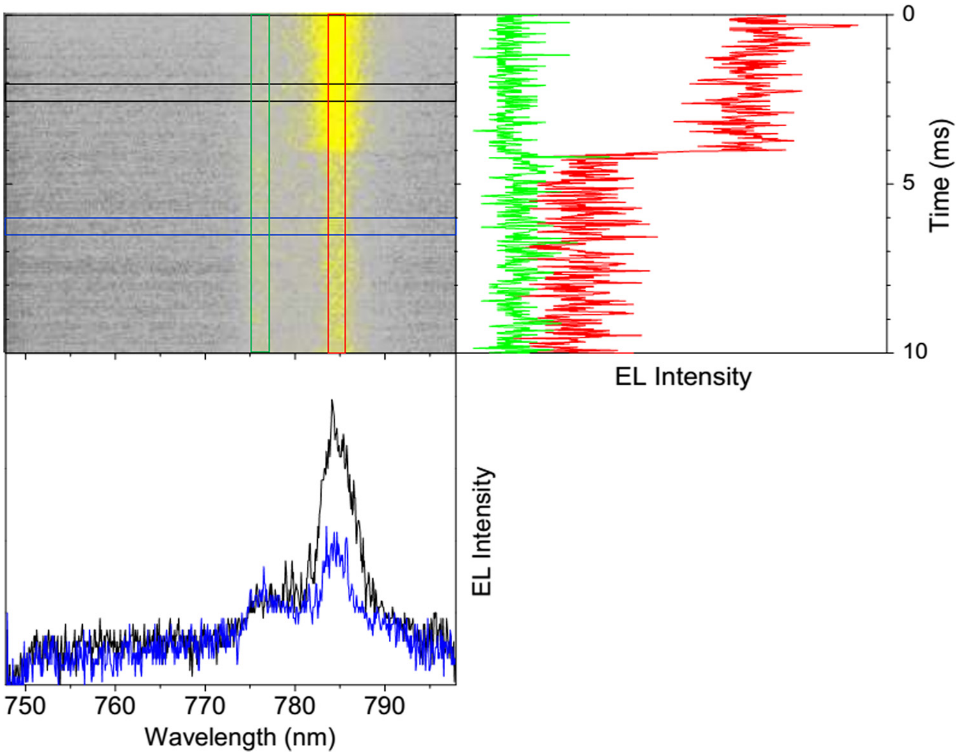

FIG. 4. Electroluminescence streak image and the correspondent transients (right side) and spectra (below) obtained at the critical bias voltage 1.7585 V where an abrupt variation of EL intensity is observed. The transients were obtained for integration interval of 2 nm around the E1-A0 (green) and E1-HH1 (red) emissions. The spectra were obtained for time integration interval of 0.5 ms at two distinct time intervals (black and blue).

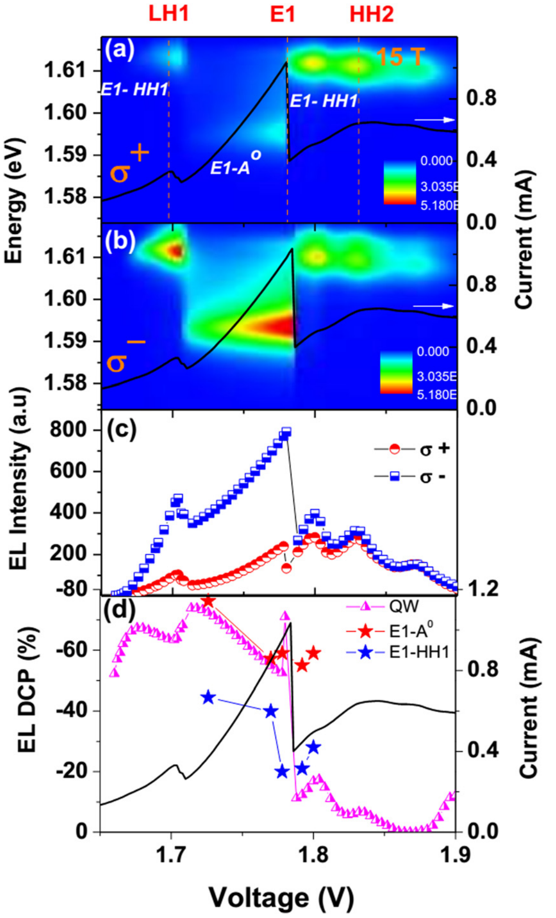

FIG. 5. (a) and (b) Color coded maps of polarization resolved QW EL emission as a function of applied bias for different values under 15 T at T = 2 K. (c) Polarization resolved of the total EL intensity as a function of bias under 15 T (d) Voltage dependence of the experimental degree of circular polarization (EL DCP) of the total QW EL and estimated values for E1-A0 and E1-HH1 emissions under 15 T and 2 K.

polarization degree from the QW emission starts from zero at B = 0 T and tends to increase with the magnetic field, but it usually shows some oscillations with B that correlate with integer filling factors of the Landau levels from the electron/hole gases.[31] Our work is focused on the polarization degree of the QW emission and the effect of its abrupt change with bias involving an impurity-related emission. Therefore, we focused on measurements under a high magnetic field (15 T), where the polarization degree is more pronounced.

For bias voltages smaller than the E1 resonant peak (<1.78 V), we obtained relatively high circularly polarization degrees (up to ∼−75%) for the total EL emission. The DCP value is rather large considering the relatively small spin splitting energies observed for the QW emission bands that are of the order of 2 meV or smaller for both emission bands. Furthermore, the spin splitting energies are approximately constant for all bias range, showing no correlation with the DCP dependence on the bias voltage. Therefore, the voltage dependence of the carrier spin polarization in the QW cannot be explained by a simple thermal occupation of the QW levels. The total DCP value shows abrupt variations that clearly correlates with the I(V) resonances, including small peaks associated with the Landau level resonances at larger voltages (>1.78 V).

In spite of the uncertainty of the DCP values obtained from the fittings, the results show that the E1-A0 transition is strongly polarized than the E1-HH1 transition for a given bias voltage, which is in fact, a rather unexpected result. This effect must therefore involve with the polarization of the injected holes, while they relax to the fundamental HH1 state or they are captured by Be acceptors. The results indicate that the occupation of the spin polarized HH1 levels reinforce a $\sigma+$ transition reducing the effective E1-HH1 DCP value. Additional investigations are, however, necessary to clarify this point. Nevertheless, the sharp variations of the DCP value for the total integrated EL emission at ∼1.70 and 1.79 V, are probably mainly related to the abrupt inversion of the dominant EL emission band at these two voltages.

IV. CONCLUSIONS

In conclusion, we observed that the EL emission from the QW comprises two distinct bands associated to the E1–HH1 transition and a recombination involving residual acceptors in the QW (E1-A0 transition). The total integrated EL emission shows a strong correlation with the I(V) resonances. Furthermore, the EL spectra shows remarkably abrupt changes at the threshold and at the resonant peak associated to the E1 resonant tunneling condition, when the dominant band switches between the E1-HH1 and the E1-A0 bands. This switch is especially abrupt at the E1 resonance, where we have observed an intriguing effect in the form of a telegraphic–random noise with sudden inversions between the two main QW emission lines when the EL emission was measured as a function of time under a constant bias voltage. The band switch is also accompanied by abrupt changes of the QW polarization degree that are consistent with the larger polarization degree of the QW recombination involving the acceptor state. Finally, our investigation revealed that a p-i-n RTD present very abrupt voltage-controlled changes of the QW EL emission including its intensity, peak energy, and spin-polarization, which may be useful for the development of future devices in spintronic applications.

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