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Phys. Rev. B **105**, 045414 — Published 18 January 2022

DOI: [10.1103/PhysRevB.105.045414](https://doi.org/10.1103/PhysRevB.105.045414)

# Spin dependent analysis of homogeneous and inhomogeneous exciton decoherence in magnetic fields

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(Dated: January 3, 2022)

This paper discusses the combined effects of optical excitation power, interface roughness, lattice temperature, and applied magnetic fields on the spin-coherence of excitonic states in GaAs/AlGaAs multiple quantum wells. For low optical powers, at lattice temperatures between 4 K and 50 K, the scattering with acoustic phonons and short-range interactions appear as the main decoherence mechanisms. Statistical fluctuations of the band-gap however become also relevant in this regime and we were able to deconvolute them from the decoherence contributions. The circularly polarized magneto-photoluminescence unveils a non-monotonic tuning of the coherence for one of the spin components at low magnetic fields. This effect has been ascribed to the competition between short-range interactions and spin-flip scattering, modulated by the momentum relaxation time.

## I. INTRODUCTION

Electronic spin in semiconductors has become a potential building block for applications in spintronics and quantum information technologies.[1, 2] This has been studied in a variety of semiconductor platforms ranging from the traditional III-V and II-VI groups to monolayers of transition-metal dichalcogenides [3–6] and significant efforts have been devoted to controlling and increasing the spin coherence time. [7–9]

Under a non-resonant optical excitation regime, photo-generated spin carriers subsequently lose energy by scattering processes followed by exciton radiative recombination. This photoluminescence (PL) is mediated by decoherence mechanisms that broaden the exciton linewidth, providing information on the time scale in which the exciton can be coherently manipulated. [10] Under an external applied magnetic field, the additional confinement in  $xy$ -plane can tune the scattering processes, also increasing the exciton spin relaxation time [11] that could lead to spontaneous coherence. [12–15] Therefore, there is an active search for understanding how and under which conditions different decoherence mechanisms of excitons are triggered. To answer these questions we investigated the relaxation process of excitons and the effect of external magnetic fields on the tuning of the spin-coherence in quantum wells (QWs). By reducing the lattice temperature and for low excitation powers, the presence of band-gap fluctuations stabilizes the carriers effective temperature at values higher than the lattice temperature. By applying a magnetic field in this regime, the exciton spin coherence can be tuned. In this case, the combination

of short-range interactions and spin-flip scattering is the leading mechanism as supported by our simulations.

## II. SAMPLE AND EXPERIMENTAL SETUP

A multiple QW heterostructure was grown via molecular-beam epitaxy on an undoped GaAs(100) substrate, consisting of twenty QWs with individual width of 55 Å and  $\text{Al}_{0.36}\text{Ga}_{0.64}\text{As}$  barriers with individual thickness of  $\sim 300$  Å - thick enough to avoid carrier tunneling between consecutive wells. Continuous-wave PL measurements were performed at temperatures ranging from 3.6 K up to 80 K by means of a confocal microscope with samples placed inside a magneto-optical cryostat (Attocube/Attodry 1000). Magnetic fields up to 6 T were applied at cryogenic temperatures. A linear polarized diode laser (PicoQuant LDH Series) was used as an excitation source ( $\lambda = 730$  nm) focused on a spot diameter of  $\sim 1$  m. A set of polarizers was used in order to identify the correspondent sigma plus ( $\sigma^+$ ) and minus ( $\sigma^-$ ) optical component emissions from the sample, which were magnified into a  $\sim 50$  m optical fiber acting as a pinhole, dispersed by a 75 cm spectrometer (Andor / Shamrock), and detected by a Silicon charge-coupled device (Andor / Idus).

## III. RESULTS AND DISCUSSION

The PL spectra at a lattice temperature of  $T_L = 3.6$  K are presented in Fig. 1(a) for various laser power densities. Here, the main electron-heavy hole emission peak at 1.615 eV, labeled as e-hh, corresponds to the  $1s$  ground state recombination. [16] At high laser power densities an emission at 1.625 eV, labeled as e-hh1, is ascribed to the

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2s exciton state. [16, 17]

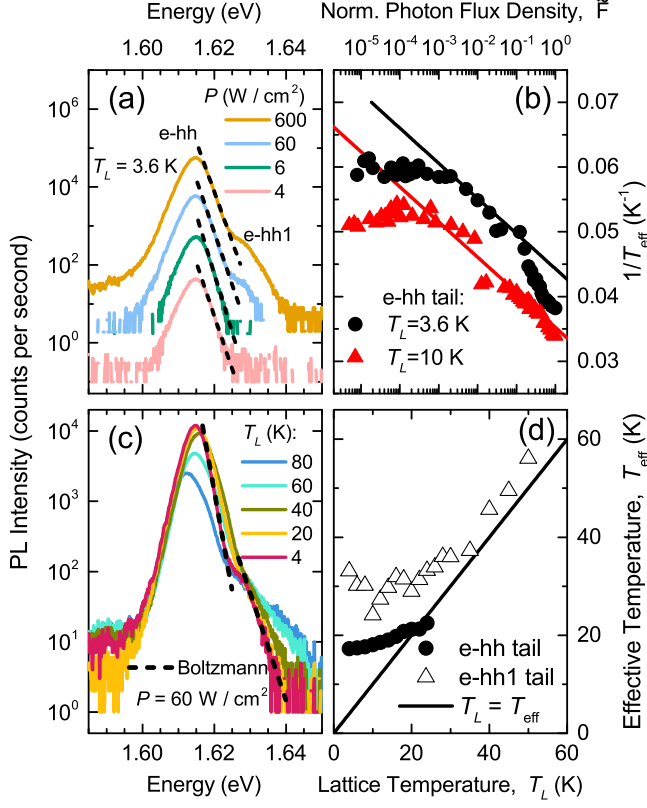


FIG. 1. (a) PL spectra emission for different laser ( $\hbar\omega_1 = 1.69$  eV) power densities at a lattice temperature of 3.6 K. (b) Inverse of the effective temperature obtained from the e-hh spectral tail versus the normalized photon flux density,  $\bar{F}$ , at a lattice temperature of 3.6 K and 10 K. Solid lines simulate the effective temperature variation produced by LO-phonon scattering,  $F \propto \exp\{-\hbar\omega_{LO}/(k_B T_{\text{eff}})\}$ . (c) PL spectra for different lattice temperatures at a constant laser power density of  $60 \text{ W/cm}^2$ . The high-energy side of the e-hh and e-hh1 emissions were fitted using Boltzmann functions (dashed lines). (d) Effective temperature obtained from the e-hh and e-hh1 spectral tails as a function of the lattice temperature. Solid line represents the case of carriers thermalization with the lattice ( $T_{\text{eff}} = T_L$ ).

Within the parabolic band approximation, the intensity of the PL signal,  $L$ , in quantum wells can be calculated as being proportional to:[18]

$$L \propto \langle u_h | \epsilon \cdot \mathbf{p} | u_e \rangle^2 D(\hbar\omega, T_e, T_h), \quad (1)$$

where  $\langle u_h | \epsilon \cdot \mathbf{p} | u_e \rangle^2$  is the dipole matrix element and

$$D(\hbar\omega, T_e, T_h) = \frac{m_r}{\pi \hbar^2} f_{\mu_e}^{\text{FD}} \left( E_g + E_1^e + \frac{m_r}{m_e} \mathcal{E}, T_e \right) f_{-\mu_h}^{\text{FD}} \left( E_1^h + \frac{m_r}{m_h} \mathcal{E}, T_h \right) H(\mathcal{E}), \quad (2)$$

being  $\hbar\omega$  the emitted photon energy and  $f_{\mu_i}^{\text{FD}}(E, T)$  the Fermi-Dirac distribution function for electron and hole

subsystems ( $i = e, h$ ) with effective temperatures  $T_e$  and  $T_h$ , and chemical potentials  $\mu_e$  and  $-\mu_h$ , respectively. These chemical potentials are measured from the valence band top. The reduced mass is  $m_r = m_e m_h / (m_e + m_h)$ ,  $H(\mathcal{E})$  is the Heaviside step function. and  $\mathcal{E} = \hbar\omega - (E_g + E_1^e + E_1^h)$  with  $E_g$  as the band-gap energy and  $E_1^{e(h)}$  the energy of the ground state for electrons (holes).

In the high-energy side of the emission spectra, the Fermi-Dirac distribution can be approximated to a Boltzmann like function,

$$f_{\mu}^{\text{FD}}(E, T) \Big|_{\frac{E-\mu}{k_B T} \gg 1} = f^{\text{B}}(E, T) = \exp\left(-\frac{E-\mu}{k_B T}\right), \quad (3)$$

and the quality of this approximation can be assessed using as reference the relative reduction of the distribution function, independently on the position of the chemical potential. It can be readily demonstrated that, the condition

$$\frac{f^{\text{B}}(E, T) - f_{\mu}^{\text{FD}}(E, T)}{f_{\mu}^{\text{FD}}(E, T)} 100\% < 10\%, \quad (4)$$

is attained once  $f_{\mu}^{\text{FD}}(E, T) < 1/11$ . Thus, the relative reduction of at least one order of magnitude below the degenerate condition, where  $f_{\mu}^{\text{FD}}(E, T) = 1$ , already guarantees staying below 10% discrepancy of the Boltzmann approximation with respect to the Fermi-Dirac distribution. Under this approximation, Eq.2 transforms into

$$D(\hbar\omega, T_e, T_h) = \frac{m_r}{\pi \hbar^2} \exp\left(-\frac{E_g + E_1^e + \frac{m_r}{m_e} \mathcal{E} - \mu_e}{k_B T_e}\right) \exp\left(-\frac{E_1^h + \frac{m_r}{m_h} \mathcal{E} + \mu_h}{k_B T_h}\right) H(\mathcal{E}), \quad (5)$$

and so the intensity becomes proportional to

$$L(\hbar\omega) \propto \exp\left(-\frac{\hbar\omega}{k_B T_{\text{eff}}}\right), \quad (6)$$

with  $(T_{\text{eff}})^{-1} \equiv m_r [(m_e T_e)^{-1} + (m_h T_h)^{-1}]$  as the e-h pair effective temperature. A discussion about the nature  $T_{\text{eff}}$  and the sources that contribute to it can be found in Ref. [19]. Although this picture points, a priori, to the possibility of the electron and hole subsystems not being mutually thermalized,[18, 19] there is no reason to assume that in this case, so  $T_e = T_h = T_{\text{eff}}$ .

Following the above-presented approximation, the e-hh high-energy spectral tail in Fig. 1(a) (black dashed lines) can be described by a Boltzmann distribution function, as indicated by Eq. 6. The value of  $1/T_{\text{eff}}$  extracted from the high-energy tail of the e-hh emission is presented in Fig. 1(b) as a function of the photon flux density defined as  $F = P/(\hbar\omega_1)$ , with  $P$  being the laser power density and  $\hbar\omega_1$  the laser energy. [20] In Fig. 1(b), the flux density has been normalized to the maximum value of the experimental range,  $\bar{F} = F/F_{\text{max}}$  for two different lattice temperatures. When the longitudinal optical

(LO-)phonon scattering is the most efficient energy relaxation process, the relation  $F \propto \exp\{-\hbar\omega_{LO}/(k_B T_{\text{eff}})\}$  is expected, with  $\hbar\omega_{LO} = 36.5$  meV being the LO-phonon energy in GaAs. [20] This is depicted in Fig. 1(b) by solid lines, in good agreement with the experimental data at high incident photon fluxes ( $\bar{F} > 10^{-3}$ ). At low optical power densities the effective temperature deviates from this trend, stabilizing at a constant value of 17 K for  $T_L = 3.6$  K, and 19 K for  $T_L = 10$  K. This points to a constriction of the LO-phonon scattering, enhancing the relative contribution of the carrier-carrier interaction which stabilizes the effective temperature. [21] As demonstrated below, this constriction is triggered in the regime where the energy band-gap fluctuations, caused by roughness at the GaAs/AlGaAs interfaces [22] play a dominant role. [23]

The PL spectra obtained with a laser power density of  $60 \text{ W/cm}^2$  are shown in Fig. 1(c) varying  $T_L$ . In this case the e-hh1 emission is well resolved and  $T_{\text{eff}}$  is displayed in Fig. 1(d) for both e-hh and the e-hh1 tails as a function of  $T_L$ . The condition of perfect thermalization with the lattice,  $T_{\text{eff}} = T_L$ , is also represented. The excited states show higher temperatures than the ground state and by increasing  $T_L$  a more efficient thermalization with the lattice is observed. We should note that for temperatures above 25 K, the e-hh tail exhibits a shoulder around 1.620 eV that hampers the extraction of reliable values of  $T_{\text{eff}}$ . The stabilization of the effective temperature at lower lattice temperatures indicates a constriction of phonon-mediated relaxation that impedes the thermalization with the lattice.

### A. Interface roughness effect

For assessing the relative role of different decoherence mechanisms, the full width at half maximum (FWHM) of the emission lines can be examined. Although the coherence loss by time-irreversible processes can be mapped by analyzing how the FWHM changes with temperature and external fields, this parameter is also affected by statistical fluctuations of the spatial variation of the light sources (excitons in this case). So the role played by the statistics of the interface roughness must be determined.

In order to assess this effect, the lattice temperature dependence of the e-hh peak position,  $E_p$ , has been displayed in Fig. 2(a) for two laser power densities along with simulations using the expression [24]

$$E_p(T_L) = E_g - \frac{\alpha\Theta}{2} \left[ \sqrt[1+p]{1 + \left(\frac{2T_L}{\Theta}\right)^p} - 1 \right] + E_1^e + E_1^h, \quad (7)$$

where  $E_g = 1.519$  eV is the GaAs energy band-gap at  $T_L = 0$  K, and  $E_1^e + E_1^h = 98$  meV is the extra energy given by the QW confinement,  $\alpha = 5.405 \times 10^{-4}$  eV/K, [25]  $\Theta \equiv \hbar\omega_{LO}/k_B$  with  $\hbar\omega_{LO} = 36$  meV for GaAs, while  $p = 2.5$  for  $60 \text{ W/cm}^2$  and  $p = 2.45$  for  $2 \times 10^4 \text{ W/cm}^2$ . Here,  $p$  is a parameter related to the

shape of the electron-phonon spectral function. [24, 26] The deviation,  $\Delta E_p$ , of the theoretical expectations and the experiment, is produced by local band-gap fluctuations provoked by interface roughness. [26] At low temperatures, excitons can be trapped into these fluctuations and, by increasing  $T_L$ , they progressively diffuse and recombine radiatively from higher energy states. [27] By increasing the laser power density [26] the band-gap fluctuations are effectively screened reducing  $\Delta E_p$ , as confirmed in Figs. 2(a), (b) and (c). These effects can be analyzed by using the model reported in Ref. 28, which describes the emission intensity as a function of the photon energy  $E$ , as

$$\phi(E) = \text{erfc} \left( \frac{E_g - E + \beta\sigma^2/k_B T_L}{\sqrt{2}\sigma} \right) E^2 \exp \left( -\frac{E - \mu_0 - \beta E_g}{k_B T_L} + \frac{\beta^2 \sigma^2}{2(k_B T_L)^2} \right), \quad (8)$$

where  $\sigma$  is the standard deviation of the local band-gap fluctuations, [26, 29] and  $\beta = [1 + (\lambda_\mu/\lambda_g)^2]^{-1/2} \in (0, 1]$  depends on the ratio of the characteristic length of the carriers transport,  $\lambda_\mu$ , with respect to the correlation length scale of the fluctuations,  $\lambda_g$ , and ponders the trapping efficiency:  $\beta \rightarrow 0$ , small-scale fluctuations (inefficient trapping) and  $\beta = 1$ , large-scale fluctuations (efficient trapping). This has been schematically represented in Fig. 2(d).

For relatively large arguments of the complementary error function,  $\text{erfc}(\xi) \approx \exp(-\xi^2)/\sqrt{\pi}\xi$ , [30] and the intensity becomes proportional to

$$\phi(E) \propto \exp \left( -\frac{E_g - \frac{\sigma^2}{k_B T_L}(1 - \beta) - E}{2\sigma^2} \right). \quad (9)$$

Thus, at low temperatures, its contribution to the FWHM is determined by the standard deviation of the gap fluctuations as  $W_\sigma = 2\sqrt{2 \ln 2} \cdot \sigma$ , while  $\Delta E_p$  can be approximated as

$$\Delta E_p(T_L) \simeq \frac{\sigma^2}{k_B T_L} (1 - \beta). \quad (10)$$

After extracting the difference between Eq. 7 and the experimental data, the value of  $\sqrt{\Delta E_p(T_L) k_B T_L} = \sigma\sqrt{1 - \beta}$  has been displayed in Figs. 2(b) and c for  $P = 2 \times 10^4 \text{ W/cm}^2$  and  $P = 60 \text{ W/cm}^2$ , respectively, showing a nonmonotonic behavior between 4 and 50 K. The corresponding experimental values of FWHM have been obtained by a Gaussian fitting of the e-hh emission as a function of  $T_L$  for the case of  $P = 60 \text{ W/cm}^2$  and displayed in Fig. 3(a). In this case a bump appears in the temperature range where the gap fluctuation effects are more evident. Since both power and temperature affect the way the fluctuations are screened, they can also tune the effective values of  $\sigma$  and  $\beta$ . Increasing power and/or temperature provokes an apparent homogenization of the fluctuations (through screening), reducing  $\sigma$

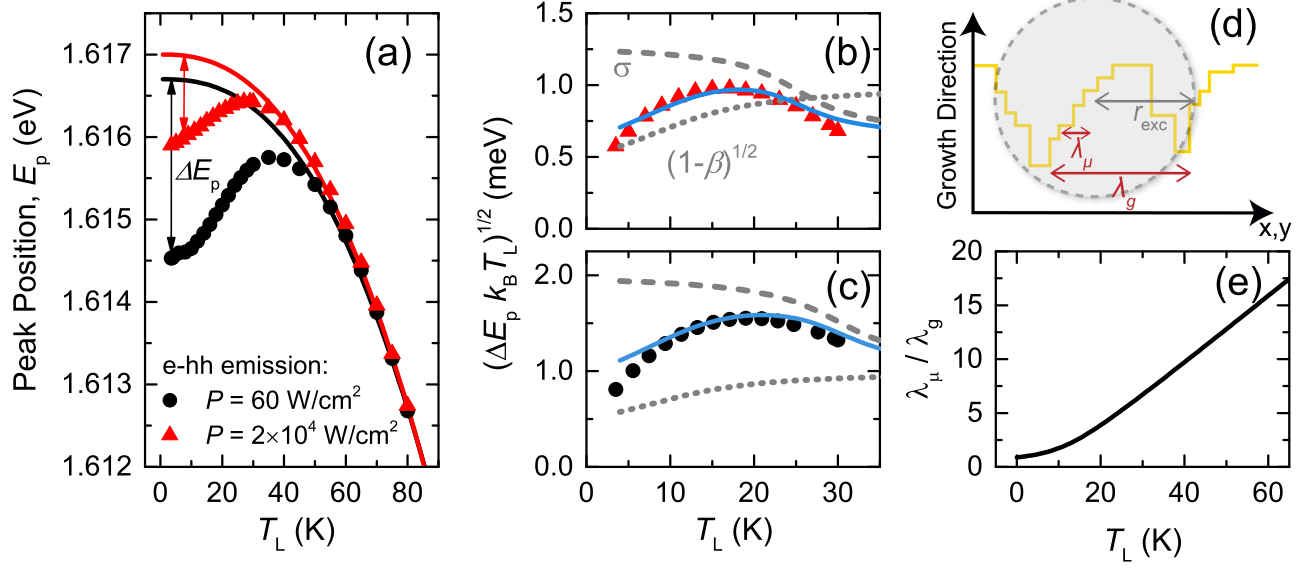


FIG. 2. (a) PL peak energy,  $E_p$ , as a function of temperature for laser power densities of  $2 \times 10^4 \text{ W/cm}^2$  and  $60 \text{ W/cm}^2$ . Solid curves represent the simulations based on Eq. 7. Temperature dependence of  $\sqrt{\Delta E_p k_B T_L} \simeq \sigma \sqrt{1 - \beta}$  for (b)  $P = 2 \times 10^4 \text{ W/cm}^2$  and (c)  $P = 60 \text{ W/cm}^2$ . Simulations (solid blue lines) were obtained by using step-like functions for  $\sigma$  (dashed line) and  $(1 - \beta)^{1/2}$  (dotted line). (d) Schematic representation of the characteristic lengths of the band gap fluctuations,  $\lambda_\mu$  and  $\lambda_g$ , as compared with the excitonic radius,  $r_{\text{exc}}$ . (e) Temperature dependence of the  $\lambda_\mu/\lambda_g$  ratio calculated from the  $\beta$  obtained in (b) and (c).

while favoring exciton diffusion that translates into a  $\beta$  decrease. By assuming  $\sigma$  and  $\beta$  as soft step-like functions with maximal  $\sigma$  and  $\beta$  for  $T_L \rightarrow 0$ , then  $\sigma$  (dashed lines) decreases by increasing  $T_L$  while  $\sqrt{1 - \beta}$  (dotted lines) grows, as illustrated in Figs. 2(b) and (c). The product  $\sigma \sqrt{1 - \beta}$  is also represented (blue solid line) reproducing the nonmonotonic behavior of  $\sqrt{\Delta E_p(T_L) k_B T_L}$  up to temperatures between 40 and 50 K. The function used to emulate  $\beta$  dependence on temperature has been the same in panels b and c of Fig. 2 and this allows extracting the expected ratio  $\lambda_\mu/\lambda_g$  that was depicted in panel e. This suggests an increased detrapping as the temperature grows.

In the case of the FWHM, displayed in Fig. 3(a), the analysis of the temperature modulation must also include the tuning of the homogeneous lifetime broadening. Thus, assuming the homogeneous broadening as a Lorentzian width,  $W_h$ , and the inhomogeneous fluctuations characterized by a Gaussian width,  $W_\sigma = 2\sqrt{2 \ln 2} \cdot \sigma$ , their relative contribution to the FWHM can be approximated as a Voigt convolution, [31, 32]

$$\text{FWHM} = \frac{W_H}{2} + \sqrt{\frac{W_H^2}{4} + W_\sigma^2}. \quad (11)$$

The homogeneous broadening can be simulated by considering different independent and additive mechanisms [23] that, for low temperatures, can be reduced to two,

$$W_H = \Gamma_0 + \Gamma_{\text{LA}}, \quad (12)$$

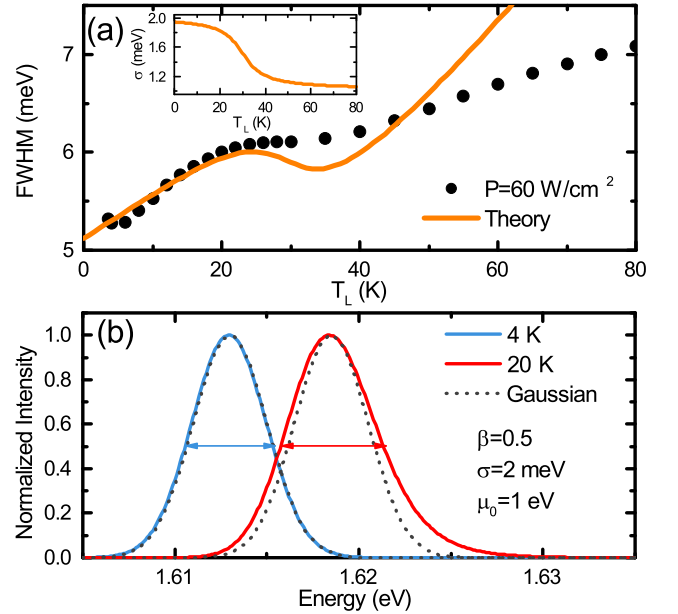


FIG. 3. (a) FWHM as a function of the lattice temperature for the e-hh emission and  $P = 60 \text{ W/cm}^2$  (dots). The simulated FWHM considering homogeneous and inhomogeneous contributions is also displayed (orange line). The  $\sigma(T_L)$  function used for the simulation has been plotted in the inset. (b) Simulated PL spectra with the same  $\sigma$  and  $\beta$  for  $T_L = 4 \text{ K}$  (blue line) and  $T_L = 10 \text{ K}$  (red line), obtained using Eq. 8 and Gaussian functions (dashed lines) according to Eq. 9.



where  $\Gamma_0$  represents the intrinsic 2D-excitonic linewidth, associated to exciton-exciton and defects scattering; [21, 23] and  $\Gamma_{LA} = 2\gamma_\alpha T_L$  arises from LA-phonon scattering. Using the function  $\sigma$  displayed in the inset of Fig. 3(a), the best simulation of FWHM at low temperatures was obtained with  $\Gamma_0 = 1.0$  meV and  $\gamma_\alpha = 0.045$  meV/K, as displayed in Fig. 3(a). Note that the simulation and the experimental values disagree for higher temperatures. As displayed in Fig. 3(b), by comparing the results using Eq. 8 and the approximation of Eq. 9, this latter does not account for the whole characterization of the spectral width modulation with temperature. A small but discernible modulation, for constant  $\sigma$ , already occurs at low  $T_L$ . As predicted in Ref. 33, the LO-phonon scattering and ionized impurities interaction do not significantly contribute to the broadening of the linewidth within the temperature range analyzed. For  $T_L < 40$  K, band-gap fluctuations broadening dominates and responds for the observed bump that can be ascribed to their effective homogenization as  $\sigma$  decreases with  $T_L$ .

### B. Magnetic fields effects

At cryogenic temperatures, phonon-assisted decoherence mechanisms are constricted. This opens the opportunity for studying the scattering processes in the presence of magnetic fields that tune the exciton size [34–36] and the exciton spin relaxation [11, 37] allowing for additional modulation of the exciton coherence. Figs. 4(a) and (b) display, respectively, the integrated intensity and FWHM of the e-hh  $\sigma^+$  and  $\sigma^-$  optical components at  $T_L = 3.6$  K, for  $P = 60$  W/cm<sup>2</sup> (top panels) and  $P = 6 \times 10^2$  W/cm<sup>2</sup> (bottom panels).

The  $\sigma^+$  component presents a monotonic dependence with magnetic field for both optical properties in Fig. 4. By increasing the magnetic field, the  $\sigma^+$  exciton population diminishes, as shown in Fig. 4(a), whereas the FWHM increases in Figs. 4(b). This FWHM increment is expected from the short-range interaction model, according to which,  $\Gamma \propto \sqrt{B}$ . [38] In contrast, the  $\sigma^-$  component in Fig. 4 exhibits a peak response in the integrated intensity and a dip in the FWHM near  $B = 1.8$  T. The peak in the  $\sigma^-$  exciton population disappears for  $T_L \geq 20$  K, as displayed by the inset in Fig. 4(a) for  $P = 60$  W/cm<sup>2</sup>.

The applied magnetic field induces the in-plane confinement of excitons, which leads to a shrinkage of the excitonic wave function and reduces the overlap with larger in-plane disorders as represented schematically in Figs. 5(a) and (b) for  $B = 0$  T and  $B \neq 0$  T, respectively. The fluctuations length-scale at the interfaces can reach widths of up to 300 Å and depths up to 2.8 Å, [22] while the excitonic Bohr radius in bulk GaAs ranges from 160 to 92 Å for magnetic fields between zero and 10 T. [39, 40] Some studies have described the modulation of the linewidth as an inhomogeneous effect where interface fluctuations of two contrasting scales are aver-

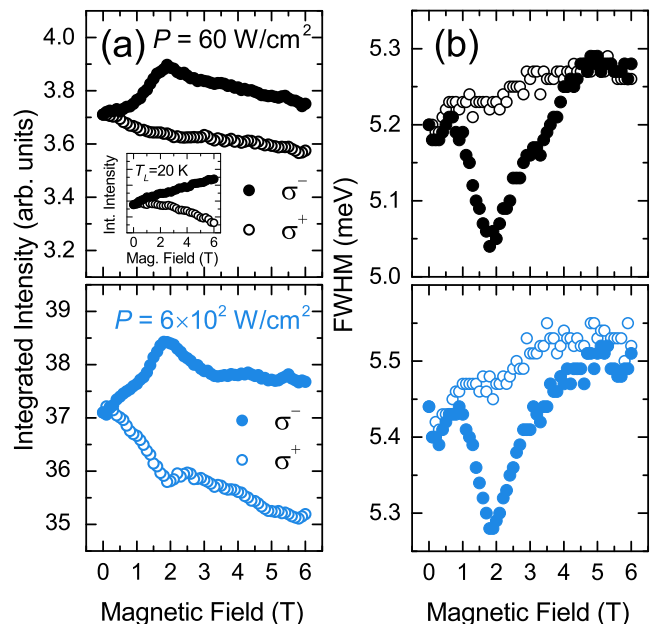


FIG. 4. Magnetic field dependence of: (a) integrated intensity, and (b) FWHM of the e-hh emission at  $T_L = 3.6$  K, obtained with  $P = 60$  W/cm<sup>2</sup> (top panels) and  $P = 6 \times 10^2$  W/cm<sup>2</sup> (bottom panels). Solid and open circles represent the responses measured from  $\sigma^-$  and  $\sigma^+$  optical component emissions, respectively. The inset in (a) shows the integrated intensity observed at  $T_L = 20$  K.

aged over the shrinking exciton radius. [35, 41] Although plausible, they cannot explain the spin modulation resolved in our experiments once the magnetic field modulation of the spin-resolved homogeneous contribution has been overlooked.

In 1991, by assuming that fluctuations caused by disorder are the dominant contribution to the line broadening, Mena et al. reported a model in Ref.[42] that extended previous theories in the absence of magnetic fields [43–46] to the magnetic field modulation. They ultimately concluded that the application of the magnetic field would enhance the value of the linewidth as the exciton radius is reduced and the disorder averaging shrinks (this is represented in panels (a) and (b) of Fig.5). However, this has been ever since in contradiction with early measurements [47, 48] and with the experimental observation of Ref.[35] where the linewidth decreases as a function of the magnetic field for 5-10 nm QWs. Although no modeling was provided to reproduce the observations in Refs.[47, 48], they do not preclude the homogeneous broadening tuning with the magnetic field of affecting the FWHM. In 1995, Aksenov et al. reported in Ref.[34], an attempt to deconvolute the homogeneous from the inhomogeneous contribution to the nonmonotonic magnetic-field modulation of the FWHM, observed in the PL emission of a GaAs QW. Here, the authors concluded, based on a phenomenological model, that homogeneous effects could be dominant at lower fields.

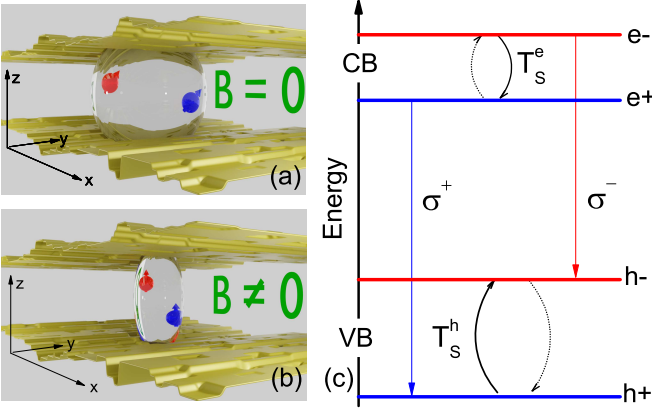


FIG. 5. Illustration of the potential fluctuations caused by roughness along with the GaAs/AlGaAs interfaces and an electron-hole pair confined in the QW with ellipsoids symbolizing the exciton size at (a)  $B = 0$  and (b)  $B \neq 0$ . (c) Representation of the spin relaxation dynamics and the optical recombination selection rules in the presence of a magnetic field.

We should note that the predictions of the theory presented in Ref.[42] were used to explain the monotonic growth of the FWHM with magnetic field finally observed in Ref.[49] (for fields up to 12 T), and also in Ref.[50] (for fields up to 42 T). Yet, these two papers neglect, a priori, any contribution of homogeneous lifetime modulation at such high fields. Later, in Ref.[35], a plausible mechanism for the inhomogeneous linewidth modulation with magnetic field was introduced, by considering the coexistence of two contrasting length scales. According to that, it is expected that the inhomogeneous linewidth, provoked by larger scale composition fluctuations, decreases with magnetic field (at least for fields up to  $\sim 15$  T) for 5-10 nm GaAs QWs, following the law

$$\sigma(B) = \sigma_0 \frac{1 - \exp[-R(B)/r_f]}{1 - \exp[-R_0/r_f]}, \quad (13)$$

where  $R(B)$  and  $R_0$  are the excitonic radius with and without magnetic field, respectively,  $r_f$  is the effective radius of the large-scale fluctuation in the plane of the QW, and  $\sigma_0$  is the linewidth at zero magnetic fields. Here,  $R(B)$  can be estimated, according to Ref.[51] as,

$$R(B) = \frac{\sqrt{2}R_0}{\left\{1 + \left[1 + \frac{3}{2} \left(\frac{e^2 R_0^4 B^2}{\hbar^2}\right)\right]^{\frac{1}{2}}\right\}^{\frac{1}{2}}}. \quad (14)$$

Note that, as expected, this magnetic field modulation leads to a monotonic decrease of  $\sigma(B)$  as the field grows, shrinking  $R(B)$  independently on the spin polarization. In this case, the exciton size shrinkage with magnetic field, represented in Figs. 5 (a) and (b), reduces the contact with larger length fluctuating interfaces [35]. However, this is still in contradiction with the modulation of the FWHM with magnetic field observed here,

that points to the need of introducing additional spin-dependent effects related to the homogeneous contributions to the linewidth.

In order to account for the polarization resolved modulation of the FWHM, we must consider the electron and hole spin splitting structure and the optical selection rules, as represented in Fig.5 (c). Note, in this picture, that the spin coherence has been assumed to be broken independently in both the conduction and valence bands ground states at rates determined by  $1/T_S^e$  and  $1/T_S^h$ , respectively. In the case of uncoupled electron-hole pairs, we can approximate their homogeneous lifetime broadening,  $W_H$ , as the sum of each component,

$$W_H^\pm = W_e^\pm + W_h^\pm, \quad (15)$$

where the homogeneous broadening for each component can be expressed as,

$$W_i^\pm = \sqrt{\frac{2}{\pi} \hbar \omega_c^i \frac{\hbar}{\tau_p^i}} + \frac{\Gamma_0^i}{1 + \left(\frac{\hbar \omega_L^i - \hbar \omega_c^i}{\hbar/\tau_p^i}\right)^2} \eta_i^\pm, \quad (16)$$

with  $\hbar \omega_c^i = \hbar e B / m_i c$ ,  $\hbar \omega_L^i = g_i \hbar e B / 2 m_i c$ , and  $i = e$  (h) labeling electrons (holes) parameters;  $m_i$  is the cyclotron effective mass,  $g_i$  the Land factor, and  $\tau_p^i$  the momentum relaxation time. The first term in Eq. 16 corresponds to the contribution of short range scattering [38] while the second term considers the spin relaxation [37, 52] inversely proportional to the spin-flip time,  $T_S^i$ , represented in Fig. 5 (c). The spin-flip rates are weighted by the thermal factor  $\eta_i^\pm = F(E_i^\pm - E_i^\mp)$  where

$$F(x) = \begin{cases} \exp\left(-\frac{x}{k_B T}\right), & x \geq 0 \\ 1, & x < 0 \end{cases} \quad (17)$$

which considers the spin energy splitting and the g-factor sign. Within this uncoupled electron-hole pair configuration, the exciton dynamics can be simplified to

$$0 = P - \frac{n^\pm W_H^\pm}{2\hbar} - \frac{n^\pm}{\tau_0}, \quad (18)$$

where  $P$  is the electron-hole pair generation rate through illumination,  $n^\pm$  is the exciton density, and  $\tau_0$  represents the optical recombination time that leads to

$$n^\pm = \frac{2\hbar P}{2\hbar + \tau_0 W_H^\pm}. \quad (19)$$

Thus, the degree of circular polarization (DCP), defined as  $DCP = (n^+ - n^-)/(n^+ + n^-)$ , becomes

$$DCP = \frac{W_H^- - W_H^+}{\frac{4\hbar}{\tau_0} + W_H^- + W_H^+} \quad (20)$$

For a quantitative analysis, the actual values of the g-factors, effective masses, and the size of the lateral (in-plane) localization of the lateral movement have been

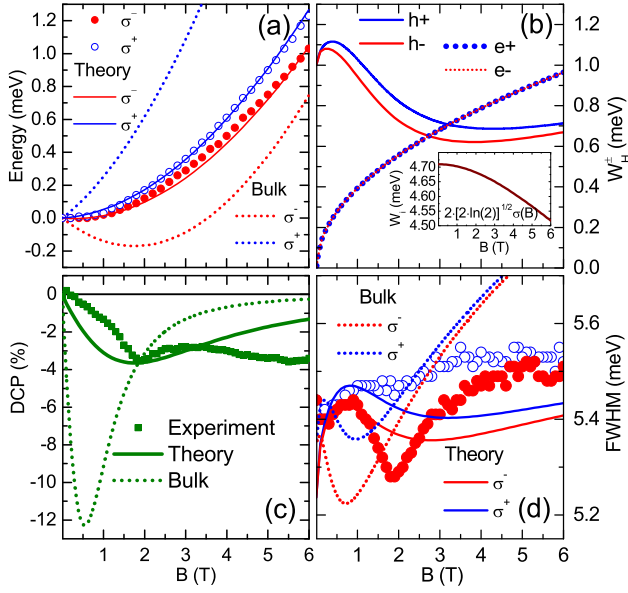


FIG. 6. (a) Experimental peak position shift as a function of the magnetic field strength for  $\sigma^+$  (blue circles) and  $\sigma^-$  (red dots) polarizations. (b) Calculated homogeneous broadening for both electrons and holes as functions of the magnetic field. The inset shows the inhomogeneous contribution. (c) Experimental degree of circular polarization (green squares). (d) Experimental values of the FWHM extracted from both circularly polarized emissions as functions of the magnetic field. In all panels, the corresponding emulated values using GaAs QW effective mass parameters are represented with solid curves, whereas those corresponding to bulk GaAs values are represented with dotted lines.

emulated within the parabolic band approximation by using the effective mass Hamiltonian [53],

$$H_{\pm}^i = \frac{\hbar^2 k^2}{2m_i} + a_1 \rho^2 \pm \frac{1}{2} g_i^* \mu_B B \quad (21)$$

for both electrons (e) and holes (h). Here,  $\rho^2 = x^2 + y^2$ ,  $\mu_B$  is the Bohr magneton, and  $\mathbf{k} = -i\nabla + \mathbf{A}e/\hbar$ , with  $\mathbf{A} = B/(2\rho)\hat{\phi}$ . The parameter,  $a_1$ , defines the strength of the in-plane confinement of the wavefunction within a site of an effective radius  $r_f^2 = \hbar/\sqrt{2a_1 m_0}$  and  $m_0$  is the bare electron mass. The ground state eigenenergy, in this case, is given by [53],

$$E_{\pm}^i = \sqrt{\frac{\hbar^4}{m_i r_f^4} + \left(\frac{\hbar \omega_c^i}{2}\right)^2} \pm \frac{1}{2} g_i^* \mu_B B. \quad (22)$$

The experimental electron-hole pair energy peak modulation with magnetic field, relative to the zero field value has been displayed in Fig. 6(a) for  $\sigma^+$  (blue circles) and  $\sigma^-$  (red dots). The data have been compared to the theoretical electron-hole pair energy calculated from Eq. 22 as  $E_{\pm}^e + E_{\pm}^h$ . By using the reported electronic structure parameters for GaAs QWs detailed in Table I, we have been able to confirm the values of the electron and hole

TABLE I. Value of the parameters used in the proposed model. Those ones extracted from the literature have their source cited.

Parameter	5.5 nm GaAs QW	Bulk GaAs
$m_e/m_0$	0.0760[54]	0.0664[55]
$m_h/m_0$	0.45[56]	$(\gamma_1 + \gamma_2)^{-1} = 0.112$ [55]
$g_e$	0[57]	-0.44[55]
$g_h$	0.7	$6.\kappa = 7.2$ [55]
$\sigma_0$	2.0 meV	2.0 meV
$R_0$	10 nm	10 nm
$\Gamma_0^e$	0	0
$\Gamma_0^h$	1.0 meV	1.0 meV
$\hbar/\tau_p^e$	0.16 meV	0.16 meV
$\hbar/\tau_p^h$	0.4 meV	0.4 meV
$\tau_0$	250 ps[58]	250 ps

g-factors. This allows assessing the size of the effective radius of the lateral confinement site provoked by the interface fluctuations as being  $r_f = 5.5$  nm, which has been used for the calculation of the inhomogeneous broadening modulation with magnetic field according to Eqs.15 and 16, and displayed in the inset of Fig. 6(b). The calculated energy peak obtained by using just bulk GaAs band parameters (included in Table I) have also been added in Fig. 6(a) as reference (dotted lines) pointing to the relevance of considering the modulation of the effective band parameters with confinement.

The corresponding values of the homogeneous width for each spin components in both conduction and valence band have been displayed in Fig. 6(b). One should note that given the negligible value of the electron g-factor and the very small cyclotron mass, the contribution of the electron spin relaxation term has been neglected and that is the reason why we can assume  $\Gamma_0^e = 0$ . The values of the rest of the parameters were set so as to get a good agreement of the calculated DCP, according to Eq. 20 with the measured values as displayed in Fig. 6(c).

Once all the parameters are determined, the corresponding FWHM for each spin component can be calculated by introducing the homogeneous contribution from Eqs. 15 and 16 along with the inhomogeneous term, defined by Eqs. 13 and 14, into Eq. 11. A reasonable agreement has been obtained with clear spin asymmetry of the linewidth in Fig. 6(d). Despite the simplicity of the parabolic approximation for the electronic structure model with relevant valence band effects, a nonmonotonic tuning of the FWHM with the magnetic field has also been obtained. The contrast, displayed in Fig. 6(d), of the expected result obtained by using bulk parameters highlights the sensitivity of this response to the band structure modulation with confinement and magnetic field. Note that, besides the eigenenergies, none of the electronic structure parameters was assumed as being spin dependent. That would lead to extra asymmetries of the homogeneous life-time broadening. Thus, the dip in the FWHM observed for the  $\sigma^-$  component is a manifestation of increased coherence with respect to  $\sigma^+$  determined by the shorter spin-flip rate described by



Eq. 16.

#### IV. CONCLUSIONS

A combined experimental and theoretical study was used to investigate exciton decoherence mechanisms in GaAs/AlGaAs multiple QWs. For temperatures below 40 K and low optical power densities, the hot carrier relaxation is affected by band-gap fluctuations produced by roughness at the GaAs/AlGaAs interfaces. These fluctuations favor carrier-carrier interactions that stabilize the effective temperature. The PL linewidth was used to characterize the exciton coherence. Both LA-phonon interaction and band-gap fluctuations affect this parameter at cryogenic temperatures. Our results demonstrate a strong modulation of these effects with temperature and optical excitation power that allowed the deconvolution of statistical contributions to the linewidth from actual decoherence mechanisms. To further tune the exciton coherence, a magnetic field was applied. At low optical power densities and lattice temperatures, observations with applied magnetic fields unveiled that spin-flip scattering and short-range interactions become the main decoherence factors responsible for the modulation of the excitonic spin-coherence. We were able to quantitatively evaluate the homogeneous and inhomogeneous

contributions to the FWHM dependence with magnetic field. Results show that the homogeneous broadening results from short-range interactions and spin relaxation processes, while the inhomogeneous contribution depends on band-gap fluctuations. Although the latter is responsible for almost 90% of the FWHM we have demonstrated that it is the homogeneous fraction that induces the spin-asymmetry and most of the non-monotonic modulation with magnetic field.

#### ACKNOWLEDGMENTS

This study was financed in part by the Coordenao de Aperfeioamento de Pessoal de Nvel Superior - Brasil (CAPES) - Finance Code 001. The authors also acknowledge the financial support of the Fundao de Amparo Pesquisa do Estado de So Paulo (FAPESP) - grants 2013/18719-1, 2014/19142-2, and 2018/01914-0, the Conselho Nacional de Desenvolvimento Cientfico e Tecnolgico (CNPq), the National Science Foundation (NSF) - grant OIA-1457888. This works was also partially funded by Spanish MICINN under grant PID2019-106088RB-C3 and by the MSCA-ITN-2020 Funding Scheme from the European Unions Horizon 2020 program under Grant agreement ID: 956548.

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