Excited exciton and biexciton localised states in a single quantum ring

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We observe excited exciton and biexciton states of localised excitons in an anisotropic quantum ring, where large polarisation asymmetry supports the presence of a crescent-like localised structure. We also find that saturation of the localised ground state exciton with increasing excitation can be attributed to relatively fast dissociation of biexcitons (∼430 ps) compared to slow relaxation from the excited state to the ground state (∼1000 ps). As no significant excitonic Aharonov-Bohm oscillations occur up to 14 T, we conclude that phase coherence around the rim is inhibited as a consequence of height anisotropy in the quantum ring.

Thanks to droplet epitaxy, quantum rings (QRs) have become an alternative means of studying the Aharonov-Bohm (AB) effect, which manifests itself by a persistent current or an oscillation of the conductance and emission energy for an external magnetic field. A delocalised wavefunction around the rim is a prerequisite for the AB effect, but the presence of localised states has been reported in a volcano-like QR arising from height anisotropy, whereby the phase coherence around the rim can be inhibited. The degree of localisation seems to depend upon the anisotropy; when this is negligible, excitonic AB oscillations begin at low magnetic fields. In the intermediate range of localisation, excitons are localised separately near the two highest rims in a QR, but the two excitons can be paired in a biexciton with sub-meV binding energy. In the case of strong localisation in a QR, no persistent current emerges unless a large magnetic field is applied. Optical AB oscillations in a QR have not been considered in the context of the localisation induced by rim height-anisotropy. This may explain why some experiments failed to observe excitonic AB oscillations in a QR. In this work, we confirm the presence of a strongly localised state in a volcano-like QR in terms of an absence of excitonic AB oscillations up to 14 T, with a small diamagnetic coefficient the polar nature and large polarisation asymmetry of biexcitons and excited excitons. These arise as a consequence of the small lateral extension and asymmetry of the strongly localised crescent-like structure in the QR.

GaAs rings were grown on an n-doped GaAs (001) substrate using a molecular beam epitaxy system with an ion getter pump, and capped with 60 nm-thick Al0.3Ga0.7As and 3 nm-thick GaAs for optical measurement. The sample was excited by frequency-doubled (400 nm) Ti:sapphire laser pulses (120 fs duration at 80-MHz repetition rate). The photoluminescence (PL) of a single QR was collected at 4.2 K using a confocal arrangement, and a time-correlated single photon counting system was used to obtain the time-resolved PL (TRPL). Magneto-PL from a single QR was also performed in a resistive DC magnet (52 mm-bore diameter) under continuous-wave Ar+ ion laser excitation (488 nm).

Although ring structures were observed in a field emission scanning electron microscope (FESEM) image of the uncapped sample (Fig.1(b)), it is known that a volcano-like anisotropic morphology is present in the QR. The height anisotropy gives rise to a crescent-shaped adiabatic potential and localisation of the potential becomes significant as the vertical quantum number is increased. For a QR with ∼20 nm-radius and ∼10 nm-height, localised states for a vertical quantum number of k = 3 can be seen at an energy close to the barrier (Al0.3Ga0.7As) bandgap. As shown in Fig.1(a), localised adiabatic potentials (∼822 eV) of the electron and the hole for a vertical quantum number of k = 3 have a crescent-like shape. An estimated ground state energy for the electron (241 meV) and the hole (60 meV) predicts a PL energy for the ground state exciton XN=1 of ∼1.812 eV in excellent agreement with the PL spectrum in Fig.1(c). It should be noted that the wavefunction contour areas should be larger than the localised potentials due to tunnelling effects.

The blueshift in the PL from XN=1 with increasing excitation power (Fig.1(e)) suggests the presence of fine structure states, where a sequential state-filling gives rise to the observed blueshift. As a consequence of asymmetry of the crescent-like adiabatic potential in Fig.1(a), the fine structure states of XN=1 within the ∼1.5 meV PL linewidth can be resolved by measuring the PL at different linear polarisation angles (Fig.1(d)). As the excitation power (Iex) is increased, two additional PL peaks emerge at low (1.808 eV) and high energy (1.822 eV) with respect to XN=1 (1.812 eV). The superlinear increase of the PL intensity (∼Iexα) was characterized in terms of the power factor (α) by integrating the PL spectrum. α ∼ 2.3 ± 0.1 and ∼ 1.5 ± 0.1 were obtained for the low and the high
FIG. 1. (a) Electrons and holes are localised in crescent-like adiabatic potentials for a vertical quantum number \( k = 3 \) \((\varepsilon_{k=3})\). (b) A single ring structure observed by FESEM. Biexciton (XX) and excited state exciton \((X^{N=2})\) emission appear near ground exciton \((X^{N=1})\) for increasing excitation power \((c)\), among which analyzer angle dependence of the PL spectrum are compared under an excitation power of 2 kW cm\(^{-2}\) \((d)\).

energy peaks compared with \( \alpha \sim 0.9 \pm 0.1 \) measured for \( X^{N=1} \) before saturation of the PL intensity. Therefore, the two additional peaks can be attributed to biexciton states (XX) and excited exciton states \((X^{N=2})\) of the localised \( X^{N=1} \), respectively, where the radial quantum number \( N \) denotes the states defined in the adiabatic potential \((\varepsilon_{k=3})\). Both wavefunctions for XX and \( X^{N=2} \) are possibly more extended and asymmetric than that of \( X^{N=1} \). However, as \( X^{N=2} \) is located \( \sim 10\) meV above \( X^{N=1} \), the range of \( X^{N=2} \) is not extended significantly \((\sim \text{few nm})\) in the contour areas of \( \varepsilon_{k=3} \) and \( \varepsilon_{h=3} \), but a node of the wavefunction must exist in the middle of the crescent structure similar to \( p \)-orbitals. This possibly results in a large polarisation asymmetry and and points to the polar nature of the wavefunction. Since XX consists of two \( X^{N=1} \)'s, the area could be nearly doubled, but shrinks due to bonding. As the observed binding energy of the XX is large \((\sim 4\) meV), this is the case of a strongly localised XX in a crescent-like structure rather than a pair of two different excitons, which are located separately at different rims\(^\text{13}\). Again, the asymmetry of the local structure gives rise to a strong polarisation dependence, where the emission energy difference of XX for perpendicular polarisations is twice \((2\Delta)\) that of \( X^{N=1} \) \((\Delta \sim 0.8\) mev) due to selection rule breaking\(^\text{14}\). On the other hand, the emission energy difference of \( X^{N=2} \) for perpendicular polarisations is \( 1.5\Delta \). Although \( X^{N=2} \) possibly has a relatively smaller wavefunction area than XX, the wavefunction shape should be very asymmetric, i.e., a node is present as in \( p \)-orbitals and is confined in the crescent-like structure. Therefore, the emission energy difference in the level spacing between the fine structure states of \( X^{N=2} \) is relatively large compared to that of \( X^{N=1} \).

As shown schematically in Fig.2(a), the underlying dynamics of TRPL can be modelled by using coupled
rate equations for $X^{N=1}$, $X^{N=2}$, and XX:

$$\frac{dN_{ex}}{dt} = g(t) - \frac{N_{ex}}{t_{ex}},$$  

$$\frac{dN_{xx}}{dt} = \gamma \frac{N_{ex}}{t_{ex}} - \frac{N_{xx}}{t_{xx}},$$  

$$\frac{dN_{x2}}{dt} = \alpha \frac{N_{ex}}{t_{ex}} - \frac{N_{x2}}{t_{x2}} - \frac{N_{x2}}{t_{x2}},$$  

$$\frac{dN_{x1}}{dt} = \beta \frac{N_{ex}}{t_{ex}} + \frac{N_{xx}}{t_{xx}} - \frac{N_{x1}}{t_{x1}} - \frac{N_{x1}}{t_{x1}}.$$  

Initially, carriers are generated in the excited state (denoted by ex) from the ground state by injection of a laser pulse ($g(t)$), which then relax quickly to $X^{N=1}$, $X^{N=2}$, and XX in a relaxation time $t_{ex}$. Because of the limited time-resolution of our TCSPC system ($\sim 50$ ps), we cannot determine $t_{ex}$; therefore, the initial population of the $X^{N=1}$ ($N_{x1}$), $X^{N=2}$ ($N_{x2}$), and XX ($N_{xx}$) are given by multiplying the intra-relaxation rate ($1/t_{ex}$) by the weight factors ($\beta$, $\alpha$, and $\gamma$), respectively, i.e., the initial carriers are distributed among $X^{N=1}$, $X^{N=2}$, and XX such that $\alpha + \beta + \gamma = 1$. While the XX dissociates into $X^{N=1}$ by radiative recombination during $t_{x2}$, $X^{N=2}$ can either relax into $X^{N=1}$ or decay radiatively to the ground state. As a result, $N_{x2}$ can be enhanced by the intra-relaxation of $X^{N=2}$ or the radiative dissociation of XX. At an excitation power near the XX onset ($1.6$ kW cm$^{-2}$) (Fig. 2(b)), the PL decay time of $X^{N=2}$ is comparable to that of XX despite the presence of the two kinds of processes in $X^{N=2}$. Therefore, the intra-relaxation between $X^{N=2}$ and $X^{N=1}$ is possibly slow, otherwise, $N_{x2}$ would show a faster decay than that of XX. When the initial weight factors ($\alpha = 0.2$ and $\gamma = 0.26$) of $X^{N=2}$ and XX are used, optimum time constants for the intra-relaxation ($\tau_{x1} = 1000 \pm 20$ ps) and radiative recombination times of $X^{N=2}$ ($\tau_{x2} = 600 \pm 20$ ps), $X^{N=1}$ ($\tau_{x1} = 500 \pm 15$ ps), and XX ($\tau_{xx} = 450 \pm 10$ ps) are obtained. These results confirm that the $N_{x1}$ enhancement is dominated by $N_{xx}$. A similar result has been obtained at the higher excitation power of $6.2$ kW cm$^{-2}$, where the PL intensity of $X^{N=1}$ is saturated. When compared with the data for $1.6$ kW cm$^{-2}$, the initial TRPL intensity of both $X^{N=2}$ and XX is increased with respect to that of $X^{N=1}$, but no significant change in the decay dynamics of $X^{N=1}$ and XX is seen (a slight change in the error bar; $\tau_{xx} = 430 \pm 10$ ps). We found again that the $N_{x1}$ enhancement is attributed to $N_{xx}$; the weight factor increase of XX ($\gamma = 0.44$) is critical whilst other constants barely change within the fitting error. Those results suggest that the relatively fast dissociation of XX fills up the $X^{N=1}$ states, then the occupied states of $X^{N=1}$ block the intra-relaxation between $X^{N=2}$ and $X^{N=1}$. It is interesting that the ratio of PL decay rate between XX and $X^{N=2}$ ($\tau_{x2}^{-1} / \tau_{x1}^{-1} \sim 1.33$) is in remarkable agreement with the ratio of the energy difference for perpendicular polarisations ($2\Delta / 1.5\Delta \sim 1.33$).

Additionally, asymmetry of the crescent-like local structure possibly gives rise to a large polarisation, whereby the optical-phonon interaction can be enhanced.

This issue can be studied in terms of the broadening of the PL linewidth as a function of temperature. As shown in Fig. 3(a), the PL intensities for all three states decrease with increasing temperature, but the PL intensity of $X^{N=2}$ becomes relatively more dominant due to thermal excitation. Additionally, both $X^{N=2}$ and XX are also dominant compared to $X^{N=1}$ in the spectral redshift and the linewidth broadening as temperature is increased (Fig. 3(b)). Given longitudinal optical (LO) phonon energy in GaAs ($\hbar\omega_{LO} \sim 36$ meV), the temperature dependent linewidth ($\Gamma(T)$) (Fig. 3(b)) is described by $\Gamma(T) = \Gamma + \alpha_T + \alpha_{opt} [\exp(\hbar\omega_{LO}/k_B T) - 1]^{-1}$, where the temperature-dependent linewidth ($\Gamma = 1.51\Gamma_0(XX), 1.27\Gamma_0(X^{N=2})$) and the coefficient of optical phonon scattering ($\alpha_{opt} = 1.81\alpha_0(XX), 1.89\alpha_0(X^{N=2})$) for XX and $X^{N=2}$ were both found to be large compared to those for $X^{N=1}$ ($\Gamma_0 \sim 0.79$ meV, $\alpha_0 \sim 132$ meV), but no significant difference was found in the coefficient of acoustic phonon scattering ($\alpha_{ac} \sim 14 \mu eV/K$) amongst all three lines $X^{N=1}, X^{N=2}$, and XX. The temperature dependence of the energy gap shift ($\Delta E(T)$), which is dominated by phonon interactions, is associated with the Bose-Einstein distribution as $\Delta E(T) = -\gamma_{ph} [\exp(T_{ph}/T) - 1]^{-1}$, where the coupling energy to the phonon bath for XX and $X^{N=2}$ ($\gamma_{ph} = 1.26 \gamma_0(XX), 1.50 \gamma_0(X^{N=2})$) was found also to be large compared to that of $X^{N=1}$ ($\gamma_0 \sim 34$ meV) with an optimum effective phonon temperature of $T_{ph} \sim 150$ K. As expected, these results confirm the stronger phonon interaction of XX and $X^{N=2}$ compared to that of $X^{N=1}$. It is also inter-
be interesting to try to induce delocalisation by either applying a larger magnetic field or by suppressing the anisotropy in the QR.

In conclusion, the presence of a strongly localised state in an anisotropic QR was confirmed, where a crescent-like asymmetric local structure gives rise to large polarisation asymmetry for localised XX and X\(N=2\) and a small diamagnetic coefficient for localised X\(N=1\). We conclude a phase coherence around the whole rim is inhibited in the case of strong localisation, resulting in no significant excitonic AB oscillations up to 14 T.

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