Tunneling blockade and single-photon emission in GaAs double quantum wells

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We report on the selective excitation of single impurity-bound exciton states in a GaAs double quantum well (DQW). The structure consists of two quantum wells (QWs) coupled by a thin tunnel barrier. The DQW is subject to a transverse electric field to create spatially indirect inter-QW excitons with electrons and holes located in different QWs. We show that the presence of intra-QW charged excitons (trions) blocks carrier tunneling across the barrier to form indirect excitons, thus opening a gap in their emission spectrum. This behavior is attributed to the low binding energy of the trions. Within the tunneling blockade regime, emission becomes dominated by processes involving excitons bound to shallow impurities, which behave as two-level centers activated by resonant tunneling. The quantum nature of the emission is confirmed by the antibunched photon emission statistics. The narrow distribution of emission energies (~10 meV) and the electrical connection to the QWs make these single-exciton centers interesting candidates for applications in single-photon sources.

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I. INTRODUCTION

The interplay between resonant tunneling and interparticle interactions in low-dimensional quantum systems gives rise to interesting phenomena in the transport of single particles. A prototype example is the Coulomb blockade, where repulsive Coulomb interaction blocks the transport through a localized prototype example is the Coulomb blockade, where repulsive Coulomb interaction blocks the transport through a localized

restricted to resonant processes leading to the excitation of individual excitons bound to shallow impurities. These states act as electrically controlled two-level systems: their quantum nature is evidenced by the antibunched photon emission statistics. The electric control and narrow distribution of emission energies make these bound-exciton emitters potential candidates for GaAs-based single-photon sources.

II. EXPERIMENTAL METHODS

The studies were carried out on the (Al,Ga)As structure illustrated in Fig. 1(a). The sample was grown by molecular beam epitaxy (MBE) on n-doped GaAs(001) substrate. The DQW consists of two 16-nm-wide GaAs QWs separated by a 4-nm-thick Al0.33Ga0.67As barrier. The DQW is subjected to an electric field $F_z$ generated by a bias voltage $V_b$ applied between a 10-nm-thick semitransparent top titanium contact [5–7] and the n-doped substrate.

The microscopic photoluminescence ($\mu$-PL) experiments were carried out in a helium bath cryostat at 4.2 K (Attocube Confocal Microscope) with positioning control provided by a piezoelectric stage. The incoming laser with a wavelength of $\lambda = 780$ nm (1.59 eV) was focused on the sample surface using an objective with numerical aperture of NA ~ 0.8. The laser energy lies below the band gap of the Al0.33Ga0.67As barriers and thus selectively excites electron hole pairs only in the GaAs QWs. The diameter of the laser spot $\phi_L = 1.2 \mu m$ sets the spatial resolution to 0.6 $\mu m$. The PL was collected by the same objective and coupled to a monochromator using either a single-mode optical fiber (for confocal measurements) or by a fiber bundle (for spatially resolved measurements). The input slit of the monochromator was selected to yield a spectral resolution of 0.08 meV. A liquid-nitrogen cooled CCD detector was used to detect the light at the output port of the monochromator. The dimension of each CCD pixel corresponds to 0.35 $\mu m$ on the sample.

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The photon autocorrelation studies were carried out using a Hanbury-Brown and Twiss setup. The PL from the shallow center was collected by the objective, filtered by a Semrock bandpass filter with a bandwidth of 810 ± 1.5 nm, and then coupled to a single-mode fiber connected to a 50/50 fiber splitter. The outputs of the fiber splitters were then sent to a pair of superconducting single-photon detectors (Single Quantum Eos). The coincidence statistics was performed by a PicoQuant Picoharp photon counting system with a time bin of 128 ps.

III. EXPERIMENTAL RESULTS

A. Exciton blockade

Figure 2(a) displays the bias dependence of the PL intensity $I_{PL}$ recorded by exciting the sample with the laser beam and integrating the emission over a 7.5-μm-long and 1-μm-wide slit across the Ti gate. The laser power is $P = 240$ nW. The PL bias map shows the typical spectral lines from DXs, IXs, and trions [8,9]. The strong trion emission signalizes the availability of free carriers. The energy and intensity of the trion line remain constant over a wide range of biases denoted as FB (for flat band) in Fig. 2(a) (i.e., for biases $V_b$ between 0.5 and 1.2 V compensating the built-in potential of the Schottky junction). Within this range, the flow of free carriers between the QWs screens the applied electric field across the DQW structure, thus inhibiting IX formation. The transition region between the FB and IX regimes is characterized by an enhanced DX emission together with a reduction of the trion intensity. The latter is attributed to the depletion of free carriers in one of the QWs as these carriers tunnel to and accumulate in the adjacent QW [8,10]. Carrier accumulation in one of the QWs also accounts for the nonlinear bias dependence of the IX energy close to the FB-IX transition [8].

More interesting spectral features appear when mapping the FB-IX transition range using a much lower excitation density, as illustrated in Fig. 2(b). Strikingly, the IX line does not smoothly “branch off” from the DX and the trion lines but only appears for $V_b < 0.25$ V, thus indicating that its formation is blocked within the bias range BR indicated in the figure. The latter results in a spectral gap between the trion energy and the onset of the IX emission given by $\Delta E_B = 5.7$ meV. Furthermore, the IX appearance at $V_b \sim 0.25$ V is accompanied by a drastic reduction of the trion intensity, which indicates that the IXs result from the dissociation of trion states. Finally, several sharp emission lines appear in the IX blockade regime BR. These lines can be grouped into
two families [labeled as DX\textsuperscript{b} and IX\textsuperscript{b} and indicated by the
dot-dashed lines in Fig. 2(b)] with bias dependence similar
to the ones for free DX and IX species, respectively. The
energies of the individual lines are typically between 1.5 and
10 meV lower than the trion emission.

The observation of multiple sharp lines in Fig. 2(b) arises
from the integration of the emission over an extended area on
the sample surface. Figure 2(c) displays a similar bias map
recorded by collecting the emission over a much smaller sam-
ple area (corresponding to the resolution limit of 0.4 μm	extsuperscript{2}),
where individual lines can be observed (labeled as D\textsubscript{1} and
D\textsubscript{2}). The energy distribution of such emission lines cover the
typical energy range of excitons bound to shallow impurities.
Natural candidates for the impurities are silicon (donor) and
carbon (acceptor), which are the most common impurities
present in the MBE growth. Interestingly, these lines mainly
appear over the narrow bias range BR and are generally not
present in the IX bias range, where their excitation should also
be energetically favorable. This behavior indicates that these
individual states are excited via a resonant process taking
place over a narrow range of biases.

The correlation between the appearance of bound exciton
and IX lines, on the one hand, and the disappearance of
trion on the other hand, suggests that the former results from
dissociation of the trion, governed by energy conservation.
The electric field dependence of the energy \(E_T\) of the different
excitonic species (\(i = DX, IX, T\) for trion) is summarized in
Fig. 2(d) [11]. The nonmonotonous electric field dependence
of \(E_{IX}(F_z)\) results from the combined effect of the QCSE and
the electric field dependence of IX binding energy \(\Delta E_{IX}\) [12].
Two trion dissociation scenarios involving resonant tunneling
and IX formation can be envisaged. For simplicity we will
consider electron trions (a similar argument applies to hole
trons). In the first, a trion dissociates via the emission of a
free carrier in the same QW (with energy equal to the band
gap energy \(E_g\)) and the tunneling of the extra electron to form
an IX, as indicated by the solid curved arrows in Fig. 2(d).
Energy conservation requires that

\[
E_T - E_{IX}(F_z) \geq E_g - E_T = \Delta_1. \tag{1}
\]

IX formation will thus be blocked within an energy range \(\Delta_1\)
below the trion energy. Alternatively, the free electron of
the previous process may form a DX in the presence of a free hole.
In this case, the condition to lift the IX formation blockade reads

\[
E_T - E_{IX}(F_z) \geq E_{DX} - E_T = \Delta_2. \tag{2}
\]

Since this process requires an extra particle, it is expected to
be suppressed at low excitation densities.

According to the calculations in Ref. [12], the binding
energy of DX, \(\Delta E_{DX} = E_{DX} - E_g\) is around 5.7 meV for the
DQW structure used here. From Fig. 2(a) we obtain \(\Delta_2 =
1.6\) meV, thus yielding \(\Delta_1 = 7.3\) meV.

The energy conservation constraint imposed by Eqs. (1)
and (2) stabilizes trions and opens a gap in the emission
spectrum of free IX species. Within the gap, trions can still
be resonantly converted into lower energy excitons bound
to an impurity, e.g., DX\textsuperscript{b} and IX\textsuperscript{b} (their energies labeled as
\(E_{DX^b}\) and \(E_{IX^b}\), respectively), indicated by the straight arrows
in Fig. 2(d). We illustrate two possible mechanisms for the
conversion. In the first case, the trion in one QW dissociates
into a free particle and a DX bound to a shallow center in the
same QW (DX\textsuperscript{b}). In the second case, one of the trion particles
tunnels across the barrier to form an IX\textsuperscript{b} bound to a shallow
center in the adjacent QW. (Note that the free particle can
also be replaced by a DX if an extra hole is available.) These
two processes yield bound species with the bias dependencies
corresponding to the ones for the DX\textsuperscript{b} and IX\textsuperscript{b} states [13,14]
indicated by the dot-dashed lines in Fig. 2(b) [e.g., both D\textsubscript{1}
and D\textsubscript{2} in Fig. 2(c) behave as DX\textsuperscript{b}].

B. Influence of laser power and temperature

Spatially integrated PL vs bias voltage \(V_b\) maps were
measured at 4.2 K for different laser excitation powers. A few
examples are shown in Fig. 3. While clear at low excitation
powers, at high excitation power the blockade is no longer
visible anymore. The blockade energy \(\Delta E_B\) is extracted as
the energy difference between T and the onset energy of the
IX detected when the bias is tuned beyond the flat-band range.
We plot $\Delta E_B$ for the lowest four excitation power $P$ in Fig. 4 and find that the blockade stays between $\Delta_1$ and $\Delta_2$.

As expected, rising temperature smears out the blockade feature, making it harder to identify, as shown in Fig. 5, all recorded at $P = 1.7 \, \text{nW}$.

**FIG. 6.** (a) Spectral PL image showing the spatial distribution of free excitons (DX and T) and excitons bound to shallow centers ($D_1$ and $D_2$). The excitons were remotely excited by a focused laser spot placed at $y = 0$. The DQW is biased at $V_b = 0.33 \, \text{V}$. (b) PL spectrum (background subtracted) at the location of $D_1$ (symbols). The superimposed lines are Gaussian fits to the resonances.

C. Bound exciton spectral properties

We now concentrate on the properties of the bound exciton states. Their spatial distribution was determined by spatially resolved PL maps recorded while biasing the structure in the BR, at $V_b = 0.33 \, \text{V}$, as illustrated in Fig. 6(a). The sample was excited by a focused laser spot at $y = 0$ and the emission recorded as a function of the distance from the excitation spot. In addition to the lines from the DX and the trion, the figure shows localized peaks from two defect centers $D_1$ and $D_2$ at 1530.4 and 1532.7 meV, respectively [the same defects as in Fig. 2(c)], which are populated by carriers diffusing from the excitation spot. The shallow centers are typically between 2 to 4 $\mu$m apart, thus yielding an areal density $n_d \approx 10^7 \, \text{cm}^{-2}$.

Figure 6(b) displays a cross section of Fig. 6(a) at the spatial position $y = 1.4 \, \mu\text{m}$ of $D_1$ (maximum emission). The spectral line shape is fitted by a Gaussian profile (red) with a full width at half maximum of $\hbar \Gamma_1 = 0.26 \, \text{meV}$. The latter is significantly narrower than the DX and trion linewidths (of 0.42 and 0.66 meV, respectively, cf. blue curves).

The linewidth $\hbar \Gamma_1$ of the bound excitons depends both on excitation power and temperature, as shown in Fig. 7 for a bound exciton labeled as $D_2$ emitting at 1.529 eV. Increasing excitation density broadens the linewidth [symbols, Fig. 7(a)]
measured at 4.2 K. For low excitation densities, the measured linewidths become determined by the spectral resolution of our monochromator, indicated by the dashed line in Fig. 7(a). The strong increase of the linewidth with excitation power points to linewidth broadening mechanism induced by charge fluctuation around the shallow center [15]. Figure 7(b) shows that an increase in temperature $T$ also broadens the bound-exciton line, which is probably due to interaction with phonons [15]. The temperature dependence was recorded for a laser excitation of 100 nW. The linewidth saturation at low temperatures is attributed to the charge fluctuation mechanism mentioned above [see also Fig. 7(a)].

By raising the sample temperature we also thermally quench the PL intensity for $D_3$, as shown in Fig. 8(a), from which we extract the activation energy $E_A$ for its dissociation [16]. The probability of thermal activation obeys the Boltzmann’s law

$$p \propto \exp\left(-\frac{E_A}{k_B T}\right). \quad (3)$$

Here $k_B$ is the Boltzmann constant. The PL quantum efficiency can be expressed as

$$\eta = \frac{1}{1 + \xi \exp(-E_A/k_B T)}. \quad (4)$$

where $\xi$ is a constant. The intensity of PL is proportional to its quantum efficiency, and thus decreases with rising temperature, as can be seen in Fig. 8(a).

We can express the temperature dependence of PL intensity as

$$\frac{1}{I_{PL}} = A + B \exp\left(-\frac{E_A}{k_B T}\right), \quad (5)$$

where $A$ and $B$ are additional constants. By fitting the data with Eq. (5) [see Fig. 8(b)], we extract $E_A = 6.5$ meV, which is comparable with the energy difference between the trion and the defect center $D_3$ of 7 meV, thus indicating that the decay is caused by thermal dissociation into the trion. Similar to the linewidth, the PL intensity saturates for $T < 5$ K.

D. Magnetic field dependence

Information about the internal structure of the bound excitons was obtained from measurements under a magnetic field $B_z$ applied along the DQW growth axis [cf. Fig. 9(a)]. The field splits the emission peak into a Zeeman doublet with energy splittings $\Delta E^{(z)}$ and average energies increasing with $B_z$. The $B_z$ dependence of the doublet energy is summarized by the blue circles in Fig. 9(b) and can be expressed as

$$E = \Delta E^{(d)} \pm \frac{1}{2} \Delta E^{(c)} = \frac{e^2}{8\mu} \rho_2^2 B_z^2 \pm \frac{1}{2} g\mu_B B_z. \quad (6)$$
FIG. 9. (a) PL spectra of D1 recorded under different magnetic fields $B_z$ applied along the growth axis (the curves are shifted vertically for clarity) showing the Zeeman splitting $\Delta E_z$ as well as the diamagnetic blueshift $\Delta E_d$. (b) Peak energies of the DX (squares) and D1 (circles) resonances as a function of $B_z$. The dashed lines are fits to Eq. (6). Due to the larger linewidths, the Zeeman splitting cannot be clearly resolved for the DXs.

Here $e$ is the electron charge, $\mu$ is the in-plane reduced excitonic mass satisfying $\frac{1}{\mu} = \frac{1}{m_e} + \frac{1}{m_{hh}}$, where $m_e = 0.0665 \, m_0$ and $m_{hh} = 0.34 \, m_0$ denote the in-plane electron and heavy-hole masses, respectively, and $m_0$ is the electron rest mass [17]. $\mu_B$ is the Bohr magneton and $g$ is the exciton Landé $g$ factor, which depends on the corresponding $g$ factors for holes and electrons [18]. The first term on the right-hand side is the diamagnetic blueshift $\Delta E^{(d)}$ resulting from the confinement of excitonic wave function by $B_z$, which depends on the spatial extent of the exciton wave function $\rho$ [19,20].

The blue dashed lines superimposed on the experimental points are fits to Eq. (6), which yield $g = 2.2$ and $\rho = 9$ nm. This value for the effective exciton radius is in good agreement with the one reported for excitons bound to shallow donors [21]. Figure 9(b) also shows the magnetic field dependence of the DX resonance (red squares). Due to the large spectral linewidth, the Zeeman splitting cannot be clearly resolved. From the quadratic dependence of the diamagnetic shifts [cf. Eq. (6)], we extract an exciton radius of 12 nm. In addition to the quadratic diamagnetic behavior, one also observes small discontinuities in the field dependence, probably due to Landau quantization levels for free carriers in the structures [10].

E. Single-photon emission

Motivated by the narrow linewidths of the bound excitons as well as their spectral separation from free exciton states, we investigate the bound-excitons’ potential as two-level systems emitting single photons. The two-level characteristic can be inferred by the saturation of emission intensity with increasing laser excitation powers, as indicated by the symbols in Fig. 10, inset. The solid line is a fit to a model for light scattered by two-level systems [22]:

$$I_{\text{PL}} = I_0 \frac{P}{P_0 + P}. \quad (7)$$

Here $I_0$ is the intensity limit approached at high input power and $P_0$ is the saturation input power, defined as the input power at which the intensity reaches $\frac{1}{2} I_0$. The saturation points to a finite number of two-level centers and also hinders the observation of bound excitons under high excitation power, as observed in Fig. 2(a).

In order to confirm the single-photon nature of the emission, we measured the second-order autocorrelation function $g^{(2)}(\tau)$ of the center D1 of Fig. 2(c) using a Hanbury-Brown and Twiss setup. The $g^{(2)}$ histogram in Fig. 10 shows the characteristic dip at zero delay $\tau = 0$, signaling photon antibunching. The solid line is a fit to delay dependence of the form $g^{(2)}(\tau) = 1 - (1/N_p) \exp(-|\tau|/\tau_R)$. The fit yields an average number of simultaneously emitted photons at zero delay $N_p < 2$, thus confirming the single-photon nature of the emission. Finally, the fitted decay time constant $\tau_R = 0.8$ ns is comparable with the values measured for self-assembled InAs/GaAs quantum dots (QDs) [23].
IV. CONCLUSION

In conclusion, we have reported the selective excitation of single excitons bound to shallow impurity centers via resonant tunneling in GaAs DQW structures. The observation of these centers becomes possible because of the blockade of carrier tunneling to form intrinsic exciton states over a range of applied electric fields. The latter hinders the formation of IXs, thus opening a gap in the emission spectrum. The PL within this gap is dominated by the dissociation of trions into individual centers by in situ fabrication of electrostatic gates. The shallow centers in GaAs DQWs are promising single-photon emitters with both optical and electrical controls.

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[11] We use electric field instead of bias to take into account screening effects.