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Vanishing fine-structure splittings in telecommunication-wavelength quantum dots grown on (111)A surfaces by droplet epitaxy

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The emission cascade of a single quantum dot is a promising source of entangled photons. A prerequisite for this source is the use of a symmetric dot analogous to an atom in a vacuum, but the simultaneous achievement of structural symmetry and emission in a telecom band poses a challenge. Here we report the growth and characterization of highly symmetric InAs/InAlAs quantum dots self-assembled on C₃ᵥ symmetric InP(111)A. The broad emission spectra cover the O (λ ~ 1.3 μm), C (λ ~ 1.55 μm), and L (λ ~ 1.6 μm) telecom bands. The distribution of the fine-structure splittings is considerably smaller than those reported in previous works on dots at similar wavelengths. The presence of dots with degenerate exciton lines is further confirmed by the optical orientation technique. Thus, our dot systems are expected to serve as efficient entangled photon emitters for long-distance fiber-based quantum key distribution.

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Semiconductor quantum dots (QDs) are expected to play a central role in quantum information networks. A noteworthy device based on dots is the solid-state single photon source, which ensures absolute security in quantum key distribution (QKD) [1]. Since QDs can confine charged carriers in nanometer-sized regions, recombination enables single photons to appear on demand, i.e., synchronously with a master clock shared in networks [2]. QKD over a 50 km commercial fiber has already been demonstrated with QD photon sources, which emitted at a wavelength of 1.5 μm [3]. The transmission distance in that work was limited purely by the absorption loss of silica fibers. Exceeding this fundamental limit requires the development of quantum link protocols, which exploit the nonlocality inherent in quantum theory. An efficient source of entangled photon pairs is a key element in the realization of such protocols, examples of which include quantum teleportation [4] and entanglement swapping [5].

The generation of entangled photons with semiconductor QDs is directly linked to the singlet configuration of two excitons (X), which form a biexciton (XX). Eventually, two photons associated with the XX-X cascade show polarization correlations independent of the choice of measurement basis, yielding quantum entanglement in the polarization state [6,7]. However, commonly studied QDs exhibit considerable fine-structure splittings (FSS) [8–12], which exclude entanglement in emitted photons [13]. Numerous attempts have been made to suppress FSS and recover the symmetry of QDs grown on conventional (001) oriented substrates [14–21]. An average FSS of ~4 μeV was also measured in GaAs QDs on nanohole template (001) substrates [22]. However, from a practical point of view, the reproducible and self-assembled growth of symmetric dots with (at least) near-zero FSS is highly desirable.

A noteworthy strategy for achieving such high QD symmetry is the application of C₃ᵥ symmetric (111) surfaces to the growth substrate, as was predicted theoretically [23]. Although QD growth in the Stranski-Krastanow (SK) mode is prohibited along the [111] axis, the use of patterned substrates [24] and droplet epitaxy [25,26] makes it possible to grow QDs on (111) substrates. Hence, a great reduction in FSS was observed in these QDs [26,27], which led to the demonstration of entangled photon emission in pyramidal QDs on patterned (111)B substrates [28], and the filtering-free violation of Bell’s inequality for droplet epitaxial GaAs/AlGaAs dots on GaAs(111)A [29]. Note that all these studies dealt with visible wavelength photons. Material challenges have meant that the development of QD sources in telecom bands has achieved less progress. The simultaneous realization of small FSS and telecom-band emission is a great challenge.

In this work, we report on the growth and characterization of telecom-band InAs quantum dots on (111)A substrates. For this purpose, we focus on the use of droplet epitaxy, which is not strain driven, thereby enabling us to choose a variety of materials and surface orientations. Though most previous works on droplet epitaxy dealt with lattice-matched systems, the versatility of this technique makes it possible to create QDs on lattice-mismatched systems targeting telecom-band emission. The successful growth of InAs/InAlAs QDs on InP(111)A has recently been demonstrated [30]. Here we use newly created QDs with a lower surface density, which allows a systematic study of FSS and the symmetry characteristics of dots. The measured FSS reveals an average value of 25 μeV, which is considerably smaller than those in previous studies on SK-grown dots at similar wavelengths [31–33]. Moreover, the presence of QDs with nearly perfect exciton degeneracy is confirmed using the optical orientation technique. Thus our source is expected to serve as a promising candidate for highly efficient entangled photon sources, which do not require the use of serious temporal gating to improve the degree of quantum correlation [34].

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The sample investigated in this study was grown by droplet epitaxy using a conventional molecular beam epitaxy apparatus [35,36]. After growing a 150-nm-thick $\text{In}_{0.52}\text{Al}_{0.48}\text{As}$ barrier layer on an InP(111)A substrate at 470 °C, we cooled the substrate to 320 °C and supplied 0.4 monolayers of indium, which led to the formation of indium droplets. Next, we supplied an As$_4$ flux of $3 \times 10^{-5}$ Torr to crystallize the indium droplets into InAs dots at 270 °C. The sample was then annealed at 370 °C and capped with another $\text{In}_{0.52}\text{Al}_{0.48}\text{As}$ barrier layer with a thickness of 75 nm.

For atomic force microscopy (AFM) analysis, an additional QD layer was grown on the top of the sample. Figure 1(a) shows a three-dimensional view of the surface, which reveals the presence of dislikle dots with $3.0 \pm 1.0$ nm in height and $38 \pm 10$ nm in diameter. The dot density is as low as $3.2 \times 10^9$ cm$^{-2}$, which makes it possible to isolate single dots using conventional microphotoluminescence techniques. Figure 1(b) shows the AFM cross sections for two example QDs. Cross sections obtained along the orthogonal in-plane directions, $[01\overline{1}]$ and $[\overline{2}1\overline{1}]$, are almost identical, which supports the high lateral symmetry in the dot shape without any elongations. This symmetric characteristic is a consequence of QD growth on (111) substrates.

For the photoluminescence (PL) measurement we used a continuous wave laser emitting at a wavelength of 705 nm for excitation above the barrier band gap. The laser light was focused on the sample using a near-infrared microscope objective with a numerical aperture of 0.65. To reduce the spot size, a hemispherical solid immersion lens with a diameter of two was positioned on the sample. In this setup, approximately 20 dots are in the excitation spot of a near-infrared microscope objective with a numerical aperture of 0.65. Nevertheless, we can isolate a single dot in spectral domain from a dot ensemble with broad size distribution. Spontaneously emitted photons were collected with the same objective, and then fed into a 50-cm focal length polychromator equipped with an InGaAs array detector. The spectrometer had a resolution of 55 μeV (0.08 nm) with a full width at half maximum (FWHM) at a wavelength of 1.3 μm. The linearly polarized PL spectra were recorded as a function of the polarization angle. With a Gaussian fit to the emission lines, we were able to determine the spectral peak shift (and the absolute value of FSS) with a resolution as high as 4 μeV. All the experiments were performed at 10 K.

Figure 2(a) shows the low-temperature PL spectrum of the dot ensembles. The PL spectrum spreads in a 1.1–1.6 μm wavelength range, which covers the O ($\lambda \sim 1310$ nm), C ($\lambda \sim 1550$ nm), and L ($\lambda \sim 1600$ nm) telecom bands. The spectrum consists of several split peaks, among which high-yield emissions are centered at $\sim 930$ meV (1333 nm) with an FWHM of 40 meV. The appearance of multiple peaks can be attributed to the different families of QDs with heights varying in monolayer steps [26]. Note that we measured the PL at a sufficiently low power where contribution from excited states can be excluded. The AFM analysis suggests that the QDs have a flat shape with heights ranging from 2 to 6 monolayers, which is consistent with the observed spectral profile.

Figure 2(b) shows the PL decay signals of the dot ensembles at wavelengths around 1.3 μm after pulsed excitation. For this measurement, we used a mode-locked Ti:sapphire laser for excitation ($\lambda \sim 785$ nm) and a superconducting single photon detector (SSPD) for detection. The decay curve reveals a single exponential with a decay time of 1.43 ns, which agrees with the theoretical decay time of spontaneous emission on the assumption that the QDs have the same transition strength as the bulk value. The similar decay times have been confirmed in telecom-wavelength QDs grown for different substrate orientations [37]. Thus, the observed PL decay is likely governed by intrinsic carrier recombination and free of any nonradiative process, as a consequence of the high crystalline quality of dots. The homogeneous linewidth, which gives the maximum limit of FSS for entangled photon emission, is thus $\sim 0.5$ μeV for our dots.

Figure 3(a) shows the typical PL spectrum of a single dot. Four emission lines are observed, and assigned, from the high-energy side, as $X$, $X'^+$, $XX$, and $X^-$, where $X'^+$ refers to positively (negatively) charged excitons. These assignments are based on the measurement of the excitation power dependence of the emission lines [Fig. 3(b)], where almost linear and quadratic behaviors were observed for $X$ and $XX$, respectively. The assignment of $X^+$ and $X^-$ is further supported by an optical orientation measurement, as described later.
The presence of dots with effectively zero FSS can be confirmed by measuring circularly polarized emission signals. In the presence of a finite FSS value, the polarization state of the emission light is expected to oscillate temporally between the left- and right-handed circular polarizations, where the oscillation period is determined by the inverse of the FSS. Therefore, in time-integrated experiments we cannot observe a high degree of circular polarization. By contrast, in the absence of FSS, circular polarization remains in time-integrated signals. Thus, the observation of circular polarization provides a sufficient condition for finding QDs with negligible FSS over a broad spectral range.

The statistical results for FSS over ~50 dots are summarized in Fig. 4(c), where the FSS magnitude is plotted as a function of X energy. A histogram of the FSS values is also shown in the Supplemental Material [40]. The FSS ranges between 70 and 3 μeV, where the minimum value is smaller than the error width of the present analysis (4 μeV, shown by the shaded region). The FSS average value is 25 μeV, which is considerably smaller than those of SK-grown QDs in the telecom band [31–33]. Note that two families of QDs with different monolayer heights are present in Fig. 4(c), as shown by the two peaks in the ensemble spectrum (broken line). Each family with a given height reveals a trend where the FSS decreases as the energy increases. This implies that high-energy QDs have a smaller in-plane size and higher lateral symmetry. The impact of the lateral-size reduction on FSS minimization was also confirmed in a previous study on FSS control by high-temperature annealing [14].

Figure 4(d) shows the direction of the polarization axis with respect to the X energy. They are randomly distributed, without showing significant correlations with the in-plane crystallographic axes. The absence of a preferential direction in the (111) plane suggests a high probability of finding QDs with negligible FSS over a broad spectral range.

The presence of dots with effectively zero FSS can be confirmed by measuring circularly polarized emission signals. In the presence of a finite FSS value, the polarization state of the emission light is expected to oscillate temporally between the left- and right-handed circular polarizations, where the oscillation period is determined by the inverse of the FSS. Therefore, in time-integrated experiments we cannot observe a high degree of circular polarization. By contrast, in the absence of FSS, circular polarization remains in time-integrated signals. Thus, the observation of circular polarization provides a sufficient condition for exciton degeneracy. The measurement principle is analogous to the well-known Hanle measurement, and was used to monitor FSS cancellation by an electric field [41]. Note that we use the 1.76-eV excitation light, which is
predominantly absorbed by the unstrained InAlAs barrier with a
direct band gap at 1.52 eV. Thus, the circularly polarized light
induces spin-polarized electron injection from the heavy-
and light-hole bands, but not from the split-off band. The maximum
degree of spin polarization is therefore limited by 0.5 based
on the assumption of fast hole relaxation. To avoid the effect
of dynamic nuclear polarization [42], we set the excitation
power at a sufficiently low level, where the average exciton
population in the dot was ~0.5.

Figures 5(a) and 5(b) show optical orientation results for a
selected dot without a detectable FSS (<4 μeV, QD1) and for
a dot with a significant FSS (~45 μeV, QD2), respectively.
The upper panels show unpolarized spectra, Iσ⁺ + Iσ−, where
Iσ⁺ (Iσ−) is the emission intensity with cocircular (cross-
circular) polarization with respect to the excitation light. The
lower panels show the differential spectra, Iσ⁺ − Iσ−, where
a pronounced positive peak appears for the X line of QD1, but
disappears for that of QD2. The positive degree of polarization
for the X line of QD1 (+4.2%) is evidence of the degenerate
exciton states. By contrast, the XX line does not exhibit a
significant polarization in both QD1 and QD2, because the
transition from XX comprises two routes with orthogonal
polarizations. The other spectral lines follow well-known
dynamics: X⁺ shows a positive degree of polarization, which is
due to spin-polarized electron injection. X⁻ shows a negative
degree of polarization, which is accompanied by the spin-flip
relaxation of electrons [43,44].

Note that only a few dots exhibit circular polarization
for the X line. A rough estimation of the probability of
finding dots with a circularly polarized X line is ~2%,
which agrees with the ratio of the natural width of our dots
(0.5 μeV) divided by the distribution of FSS (25 μeV).
Note that this probability is smaller than that previously
observed for GaAs/AlGaAs QDs on (111)A (~5%) [29].
The small probability of telecom-wavelength dots reflects
the relatively long emission lifetime as compared with that
of visible-wavelength dots (560 ps for GaAs dots). Optical
orientation therefore serves as an efficient way to select dots
suited for entangled photon generation.

The advantage of using (111) substrates over using (100)
substrates is that we can exclude in-plane asymmetry on
average. Nevertheless, shape variation inherent in the dot
self-assembly gives rise to small but significant FSS in the
majority of dots. Moreover, recent theoretical attempts suggest
the influence of atomic-scale symmetry breaking on the emer-
gence of FSS [45]. Such microscopic asymmetry comes from
interfacial randomness at heterosurfaces and compositional
fluctuations inside and outside dots. It is noteworthy that the
distribution of the measured FSS in our dots is smaller than that
theoretically predicted for telecom dots on (100) surfaces, in
which a perfectly symmetric shape was assumed [46]. This
implies that the [111] grown dots are more stable against
microscopic disorder than the [100] grown dots.

We attribute the further reduction of FSS in the [111]
grown dots compared with that of conventional [100] grown
dots to two mechanisms. First, owing to the high surface
stability of the (111) plane, the dots have atomically flat
surfaces, which were demonstrated by transmission electron
microscopy analysis [30]. The smooth and abrupt interface
also leads to the observation of distinct spectral multiplets
in ensemble spectra [Fig. 2(a)]. Thus, we expect the effect
of interfacial randomness on FSS to be greatly suppressed
compared with SK-grown (100) dots. Second, in zinc-blende
compound semiconductors, the piezoelectric field direction is
along the [111] polar axis, which coincides with the vertical
growth direction in our system. Thus, a strain field does not
induce any great reduction in lateral symmetry. The
(111) surface is thus an ideal substrate for the growth of
symmetric dots, where both geometrical (shape) symmetry
and atomic-scale symmetry are well conserved.

In summary, we have presented measurements of mini-
mized FSS in telecom-wavelength InAs QDs on an InP(111)A
substrate prepared by droplet epitaxy. Polarization-resolved
PL measurements were performed to examine the FSS dis-
tribution. Resolution-limited splittings (smaller than 4 μeV)
were confirmed. The random distribution of the polarization
axis made it possible to find symmetric dots over a wide
spectral range. Thus our InAs/InAlAs dots on (111) substrates
can play a crucial role in quantum information processing as
an efficient entangled photon source that can work in telecom
fiber networks.

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