Size dependent line broadening in the emission spectra of single GaAs quantum dots:
Impact of surface charges on spectral diffusion

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Making use of droplet epitaxy, we systematically controlled the height of self-assembled GaAs quantum dots by more than one order of magnitude. The photoluminescence spectra of single quantum dots revealed the strong dependence of the spectral linewidth on the dot height. Tall dots with a height of ~30 nm showed broad spectral peaks with an average width as large as ~5 meV, but shallow dots with a height of ~2 nm showed resolution-limited spectral lines (≤120 µeV). The measured height dependence of the linewidths is in good agreement with Stark coefficients calculated for the experimental shape variation. We attribute the microscopic source of fluctuating electric fields to the random motion of surface charges at the vacuum-semiconductor interface. Our results offer guidelines for creating frequency-locked photon sources, which will serve as key devices for long-distance quantum key distribution.

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Introduction. Numerous photonic applications using semiconductor quantum dots rely on the discrete and delta-function-like density of states [1]. However, various single dot spectroscopy studies have confirmed significant line broadening in the photoluminescence spectra that is normally much broader than the transform limited width determined by the spontaneous emission rate. The line broadening mechanism is commonly attributed to spectral diffusion, where the transition frequency randomly changes through the fluctuation of a local electric field in the vicinity of dots [2–6]. The fluctuating spectral line becomes integrated into a relatively broad peak thanks to the long time scales of signal integration compared with those of environmental motion.

Some progress has been made in studying the short time scale dynamics of the spectral fluctuation. Photon correlation measurement can elucidate spectrally diffuse photoluminescence with subnanosecond characteristic times [6, 7]. The correlation functions routinely show monoexponential decays, which implies efficient coupling between a single dot and a small number of environment configurations. In contrast, resonant fluorescence measurements reveal broad-band noise spectra in the 0.1 Hz to 100 kHz range, where contributions can be expected from a large number of environment configurations [8]. Photon counting statistics of resonant fluorescence further reveal the Gaussian distribution of the random environmental shifts, which might be a consequence of the central limit theorem adopted for a large ensemble [9]. By contrast, high-resolution Fourier transform measurement confirms a motionally narrowed Lorentzian lineshape associated with rapid environmental fluctuation [10]. More recent work on field-effect devices identifies charge traps at the barrier/well interface [11] or impurity centers [12] as a dominant field source. Thus, the timescales and magnitudes of spectral diffusion vary greatly depending on the sample and the measurement conditions. We still lack a global understanding of the microscopic mechanism of spectral diffusion, however it is needed for developing frequency-locked photon sources as basic elements in long-distance quantum key distribution, e.g., quantum repeaters for extending the key transmission distance.

In this work we experimentally analyze the dependence of quantum dot morphology on environment-mediated spectral broadening. For this purpose we focus on GaAs quantum dots grown by droplet epitaxy, which enables us to continuously control the quantum dot height by more than one order of magnitude. The morphology tunability contrasts with that of traditional quantum dot growth using the Stranski-Krastanow mode, where the dot profile is essentially fixed by strain relaxation and surface energies. The spectral linewidth of a single dot emission depends strongly on the dot height. The measured height dependence agrees with that of Stark coefficients along the growth direction (normal to the sample surface). We attribute the source of the electric field fluctuation to the change in the microscopic configuration of surface charges at the vacuum-semiconductor interface [13, 14]. Thus, morphology engineering is an alternative route to achieving narrower emitter linewidths without the need for feedback techniques to suppress spectral fluctuation [15, 16].

Experimental procedure. GaAs quantum dots were self-assembly grown in Al0.3Ga0.7As by droplet epitaxy on semi-insulating GaAs(100) substrates [17, 18]. These dots are free from strain thanks to the negligible lattice mismatch between GaAs and Al0.3Ga0.7As. After the
growth of a 100 nm Al$_{0.3}$Ga$_{0.7}$As layer, different amounts of gallium ($\theta_{Ga}$) with 1.5, 2, 3, 5, 7.5, or 10 monolayers (ML) were deposited at 0.5 ML/s and 200 °C. This step enabled the formation of gallium droplets. Then, an As$_4$ flux was supplied at $2.5 \times 10^{-4}$ Torr and 200 °C, and the gallium droplets were fully crystalized to GaAs dots. Note that the As$_4$ flux was roughly two orders of magnitude higher than that used for the self-assembly of quantum ring structures [19].

After the dots were grown, the sample was annealed at 400 °C in situ (under a weak As$_4$ supply) for 10 min, and partially capped with a 20 nm Al$_{0.3}$Ga$_{0.7}$As layer. The temperature was then increased to 580 °C, while the capping continued with a 30 nm Al$_{0.3}$Ga$_{0.7}$As layer followed by a 10 nm GaAs layer. GaAs dots on a 2 ML Al$_{0.3}$Ga$_{0.7}$As layer were additionally grown on the top of samples for atomic force microscopy (AFM) analysis. Finally, rapid thermal annealing was carried out at 800 °C for 4 min in a N$_2$ atmosphere. All the samples with different amounts of $\theta_{Ga}$ exhibited well-defined dots; see AFM top views in Supplementary Fig. 1. The dot density depended only slightly on $\theta_{Ga}$ from $1.8 \times 10^{10}$ cm$^{-2}$ (1.5 ML) to $1.2 \times 10^{10}$ cm$^{-2}$ (10 ML). Thus, we assume that the volumes per dot are nearly proportional to $\theta_{Ga}$.

We used a continuous-wave laser that emitted at a wavelength of 532 nm as an excitation source. The laser illumination generated photocarriers in the Al$_{0.3}$Ga$_{0.7}$As barrier. The excitation polarization was set to be linear in order to avoid a spectral shift of nuclear origin [20, 21]. Our confocal setup combined an objective lens with a numerical aperture of 0.55 and a hemispherical solid immersion lens (SIL) with a refractive index of two.

Photoluminescence spectra. Figure 3(a) shows the spectra of a large ensemble of quantum dots. They were measured using long-focus optics. The spectral peaks at $\sim$ 1.51 eV originate from impurity-bound excitons in the GaAs substrate. Signals associated with quantum dots are observed at 1.85, 1.8, and 1.67 eV in the 1.5, 2, and 5 ML samples, respectively. The emission peak,
FIG. 3. (Color online) Comparison of (a) photoluminescence spectra of a large ensemble of GaAs quantum dots, and (b) those of a small number of dots selected using a micro-objective setup. Therefore, shifts to a lower energy side with increasing droplet volume. The 7.5 ML sample shows a relatively narrow peak at 1.55 eV, which is close to the bulk band gap of GaAs. Figure 3(b) shows the emission spectra of a small ensemble of quantum dots that were spatially selected using a micro objective setup. The spectra of both the 1.5 and 2 ML samples consist of sharp lines, whose linewidths are close to, or less than, the instrumental response of our spectrometer. There are around 70 spectral lines, which is approximately three times the expected number of dots inside a focusing spot. The discrepancy is reasonable because each dot is able to generate three to four emission lines through the formation of different types of charged/neutral exciton complexes [24]. In contrast, the 5 and 7.5 ML samples exhibit relatively broad peaks that dominate the emission signals at energies below 1.75 eV. Note that a few sharp lines are also observed at energies higher than 1.8 eV, as found with the spectral lines of the 1.5 and 2 ML samples. Thus, the broad peaks for low-energy dots and narrow peaks for high-energy dots are not sample-specific signatures, but universal size-dependent behaviors.

Figure 4 shows linewidth statistics as a function of emission energy. Here we evaluate the FWHM of all the spectral lines by fitting without distinguishing between the neutral and charged transitions. Such treatment is sufficient to clarify the general trend of size-dependent broadening, since the difference between the neutral and charged exciton linewidths is much smaller than the observed dot-to-dot variation, as confirmed previously [25]. The compiled statistics demonstrate a clear correlation between line broadening and emission energy. The smooth transition over the data points of different samples confirms that the linewidth reaches several meV for tall dots, and decreases monotonically to the resolution limit with decreasing dot height. The similar linewidth dependence on emission energies has recently been reported for polar nitride quantum dots [26].

**Environment-induced line broadening.** The effect of the environment on spectral shifts is twofold. First, hyperfine coupling between an electron and nuclei induces the Overhauser field, which acts as an effective magnetic field in the tens of mT range [27]. Second, a charge distribution in the vicinity of dots induces a local electric field. However, the effect of nuclear fluctuation on line broadening is considered to be negligible at least in the present samples, because a typical value for a nuclear field is 10 mT, which corresponds to a spectral shift of 0.25 µeV for GaAs [21]. This is much smaller than the observed linewidth, which reaches several meV. Thus, the following discussion deals only with the effect of electric field fluctuation on line broadening.

A local electric field has various microscopic origins. A common example of a field source is charge particles trapped in impurities or defects. However, their densities are normally very low in samples grown with molecular beam epitaxy (MBE, \(\sim 10^{14} \text{ cm}^{-3}\)). Hence, it is difficult for the bulk impurities or defects to realize line broadening that are comparable to the measured spectra, as also discussed later. Despite the charging and discharging of trapping centers close to dots, here we propose the fluctuation of charge carriers trapped by the vacuum-semiconductor interface as a field source.

The formation of surface states, which trap charge carriers, is linked to the presence of electronically active defects at the vacuum-semiconductor interface [28]. It is known that the surface state density depends on orientations and chemical treatments, and reaches \(10^{14} \text{ cm}^{-2}\) for a naturally oxidized GaAs(100) surface [29]. Charge carriers are efficiently trapped by the surface states, and induce a local electric field normal to the surface on average. The phenomenon also serves as the origin of band bending and Fermi-level pinning [30]. When the sample is optically excited, some of the photo-injected carriers recombine with surface charges, and others occupy different surface states. Consequently, the microscopic charge arrangement changes randomly, which gives rise to field fluctuation and spectral broadening through time integration. The effect is orientation dependent, and taller
dots become more sensitive to the induced field.

**Size-dependent Stark coefficients.** Qualitative explanation of the measured line broadening is based on the derivation of Stark coefficients and the simulation of field fluctuation. The Stark shift $E_S$ of a single-particle level is described by the second-order perturbation of the interaction Hamiltonian, i.e.,

$$E_S = \sum_{n \geq 2} \frac{|\langle \psi_1 | eFz | \psi_n \rangle|^2}{E_1 - E_n} = (eF)^2 \sum_{n \geq 2} \frac{Z_{1n}^2}{E_1 - E_n}, \quad (1)$$

where $F$ is an electric field, and $Z_{1n} = |\langle \psi_1 | z | \psi_n \rangle|$ is a dipole moment between the ground state and the $n$th excited state along a direction parallel to the field. The above equation demonstrates the size dependence of Stark shifts, where the dipole moment is proportional to the confinement length $L$, the energy denominator is scaled by $(\pi^2 \hbar^2 / 2m) L^{-2}$, hence the Stark coefficient is enhanced as the fourth power of the effective dot size along a built-in field.

Figure 5(a) shows the field dependence of spectral shifts calculated for a 12-nm-high GaAs dots. The lines show the analytic dependence for a model based on infinite-potential quantum boxes with $m_e^* (m_h^*) = 0.067 (0.5)$, and the circles are the results obtained with a more precise model, which takes account of the finite GaAs/Al$_0.3$Ga$_0.7$As potential and the effect of valence-band mixing in terms of four-band $k \cdot p$ perturbation. Both models exhibit parabolic dependence, as expected. Enhanced shifts in the finite-potential dot with respect to the infinite-potential dot arise due to the extended wave function. These results imply that an energy shift as large as 1 meV, which is a typical linewidth in the measured spectra, requires a field strength of the order of 10 kV/cm, which is expected at a position only ~8 nm from a point charge. Accompanying impurities or defects in such close proximity to dots is fairly uncommon for MBE grown samples. This is why we have excluded bulk trapping centers and proposed surface charges as a field source.

**Simulation of field fluctuation.** We evaluate the field fluctuation using a Monte Carlo simulation, where an electric field is induced by randomly positioned charge particles in a flat layer. Figure 5(b) shows the field strength distribution at a point 50 nm from the surface (dielectric constant $\epsilon = 13$). This condition reproduces the geometry of our structure. We found that the field changes randomly with different charge arrangements. The statistics yields a mean field strength $F_0$ of 7 kV/cm and a standard deviation of 1.8 kV/cm for a charge density of $1 \times 10^{11}$ cm$^{-2}$. The validity of this simulation is confirmed by the agreement between the observed mean strength and the value predicted for a uniform charge sheet, $F_z = \sigma / 2 \pi \epsilon \epsilon_0 \approx 6.97$ kV/cm, where $\sigma$ denotes a charge density. We performed the simulation for different values of $\sigma$, and found that the magnitude of field fluctuation $\Delta F$ is nearly proportional to $\sqrt{\sigma}$.

We assume that $\Delta F$ transfers proportionally to line broadening $\Delta E_S$, i.e.,

$$\Delta E_S \approx \Delta F \frac{\partial E_S(F)}{\partial F} \bigg|_{F=F_0} = E_S(F_0) \frac{2\Delta F}{F_0}. \quad (2)$$

The substitution of Eq. 1 into Eq. 2 yields the line broadening dependence on the transition energy of dots. Figure 6 compares the experimental linewidths and the calculated spectral fluctuation for different charge densities. There is fairly good agreement between the experimental widths and calculated broadening when the charge density is of the order of $10^{11}$ cm$^{-2}$, which is a reasonable value [31].

**Conclusions.** Spectral diffusion in the photoluminescence of single quantum dots is an interesting phenomenon that bridges microscopic random dynamics and macroscopic optical response. Here we studied morphologically controlled GaAs quantum dots grown by droplet
epitaxy to understand the source of environmental fluctuation, and demonstrated the impact of fluctuating surface charges on dot line broadening.

From a technological point of view, however, the line broadening phenomenon is unfavorable for practical applications of quantum dots to photon emitting devices. The present results suggest several ways to engineer spectral broadening. First, we expect to suppress line broadening by creating dots with a sufficiently low aspect ratio that are robust as regards a random electric field normal to surface. Second, we expect to achieve narrower spectra by embedding dots more deeply in the barrier matrix, where the effect of random charges at the surface would be effectively smoothed out. Line broadening is considered to decrease with the inverse square of the dot-surface distance. Finally, the use of a substrate with a chemically stable surface, such as a gallium terminated (111)A surface, and/or defect passivation technologies are another potential route by which to reduce surface charge fluctuation.

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