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Unconventional quantum-confined Stark effect in a single GaN quantum dot

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We show that a peculiar excitonic effect and a confining potential in self-assembled hexagonal GaN/AlN quantum dots produce an unconventional quantum-confined Stark effect. In contrast to the conventional quantum-confined Stark shift, the emission line from a single GaN dot under the applied electric field perpendicular to the growth direction blueshifts nearly symmetrically with respect to the direction of the field. The field dependence of the emission lines is reproduced in a charge self-consistent effective mass calculation, taking into account strain, piezoelectric charge, and pyroelectric charge. The unconventional blueshift is attributed to a significant variation of the exciton binding energy, made obvious by a cancellation between the energy shifts of electron and hole confined states.

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Quantum-confined Stark effects, the effects of an electric field on confined carriers, have been studied extensively for quantum wells and dots both theoretically and experimentally.[1–10] Many important and interesting phenomena have been observed for the Stark effects. For example, Stark shift has been used to tune the energy levels of a quantum dot[1] and a quantum molecule.[2] The fine tuning and manipulating of the electronic states via the Stark effect are essential for applications to quantum information technology.

Most of the experimental and theoretical studies have predicted and observed a redshift of the emission line with an increasing electric field because of the polarization of electron-hole pairs. An exception is the blueshift, which appears as a part of an asymmetric shift due to a partial compensation of the built-in dipole moment along the field direction. Contrary to the general features, we find in this Rapid Communication a nearly symmetric blueshift of an emission line from a single GaN/AlN quantum dot. A self-consistent calculation reproduces the unconventional shift. Spatial separation of the electron and hole due to the piezoelectric field found in nitride dots causes different confinements for the electron and hole, which leads to a cancellation of their energy shifts. Then, the exciton energy shift becomes a direct observation of a rapid reduction of the exciton binding energy. A similar binding energy reduction was found in SiGe/Si quantum wells in a very weak field regime.[3] Our observation of the Stark shift in the zero-dimensional system is important for the basic physics of excitons in a quantum-confined system, because the result provides a new type of quantum-confined Stark effect where the excitonic effect is made prominent by the peculiar three-dimensional confinements for the electron and hole.

Self-assembled GaN quantum dots[11,12] are becoming a subject of increasing interest for their potential technological applications from short-wavelength light-emitting devices such as free-space single-photon sources[13] to a quantum information processing device based on large electronic dipoles in nitride dots.[14,15] The observed Stark effect is promising also for such GaN-based applications in quantum information technology, because it provides a way to largely vary the polarization of emitted photons and the electronic dipoles.

The sample investigated was grown by metal-organic chemical-vapor deposition on an *n*-doped 6H-SiC substrate. After a growth of a 100-nm-thick undoped AlN buffer layer, GaN quantum dots and a 20-nm-thick AlN capping layer were grown. The dots have a density of $\sim 1\times 10^{10}$ cm^{-2}. To apply a lateral electric field, we processed interdigital gate electrodes (Al/Au) with a 200-nm spacing on top of the sample [Fig. 1(a)]. Microphotoluminescence measurement was performed on the sample at 4 K with a 266-nm continuous-wave laser. The excitation power was limited to 2×10^{2} W/cm^{2}.

Figure 1(b) shows a series of microphotoluminescence spectra. An isolated emission peak[16] is attributed to a single GaN dot, which is observed on a broad background emission from AlN bulk.[11] The photoluminescence emission peak blueshifts for both directions of the electric field applied perpendicular to the growth direction. Its intensity rapidly quenches with the field. The field dependence of the spectra is roughly symmetric [Fig. 1(c)]. A weak asymmetry is attributed to a slight contribution of a vertical electric field. A vertical electric field applied on GaN dots produces a strongly asymmetric shift that follows the conventional Stark effect formula.[17] A strong vertical built-in field of piezoelec-

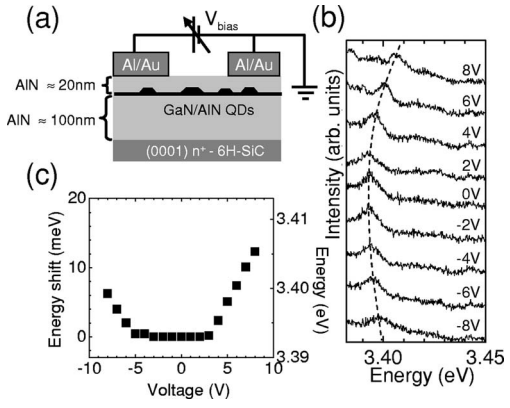

FIG. 1. (a) Schematic of the sample structure used to apply the electric field. (b) Microphotoluminescence spectra for applied voltages between −8 and 8 V. (c) Emission energy shift as a function of the applied bias voltage.

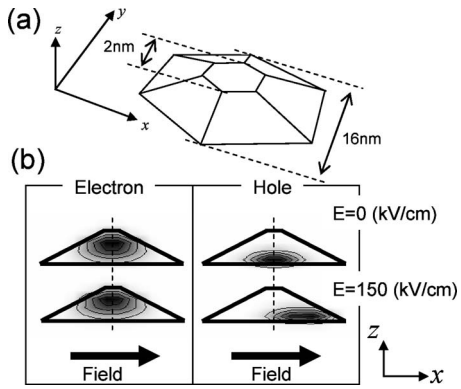

FIG. 2. (a) Schematic of the GaN/AlN dot used for the calculation. (b) Calculated probability density distribution for the ground electron and hole states.

tric and pyroelectric origin makes the linear dipole-moment term predominant, resulting in an almost linear field dependence of 0.17 meV/(kV cm^{-1}) in our dots.[18] A two-dimensional finite-element analysis solving Poisson's equation shows that our sample geometry yields a lateral electric field of about 100 kV/cm in the center between two gate fingers at 5 V. Residual doping was not considered for simplicity. Ten percent of the lateral field can produce the asymmetric Stark shift of about 2 meV, which is consistent with the result. Thus, except the slight vertical field contribution, we have observed the symmetric blueshift, which clearly deviates from the conventional Stark effect. We have observed similar blueshifts in five dots with nearly the same emission energy.

Single exciton states and their Stark shift for our GaN/AlN dots were calculated based on structural information obtained from atomic force microscope and transmission electron microscope (TEM) studies for similar dots.[19] Although there have been several theoretical studies on the lateral quantum-confined Stark effect[4,5] and experimental reports in GaAs/AlGaAs fluctuation dots,[6] InAs dots,[7,8] CdSe/ZnSe dots,[9] and InGaN/GaN dots,[10] symmetric blueshift has not been reported. In theoretical publications of electronic properties of hexagonal GaN/AlN quantum dots,[20–22] the effect of the external electric field has not been discussed. We have used a single band effective mass Hamiltonian to calculate the ground states of single-particle states including strain, piezoelectric charge, pyroelectric charge, electron-hole Coulomb interaction, and external electric field. The exciton energies are obtained variationally in an iterative Hartree scheme.[23]

The quantum dot is modeled with the shape of a truncated hexagonal pyramid with a bottom diameter of 16 nm, a top diameter of 2 nm, and a height of 4 nm [see Fig. 2(a)]. The strain distribution in and around the dot is obtained from a continuum elasticity theory implemented by a finite-element method. The piezoelectric polarization induced by the strain and spontaneous polarization found in wurtzite GaN and AlN produce electrostatic potential V_p. These are included in the electron (hole) Hamiltonian ($H_{e(h)}$).[21] In the presence of external electric field ($\mathbf{F}$) the Hamiltonians read

$$H_e = U_c - eV_p + e\mathbf{F}\cdot\mathbf{r} + \frac{\hbar^2}{2}\left(k_x\frac{1}{m_c^{\parallel}}k_x + k_y\frac{1}{m_c^{\parallel}}k_y + k_z\frac{1}{m_c^{\perp}}k_z\right) + a_c^{\parallel}(e_{xx}+e_{yy}) + a_c^{\perp}e_{zz}, \quad (1)$$

$$H_h = U_v - eV_p + e\mathbf{F}\cdot\mathbf{r} + k_x(A_2+A_4-A_5)k_x + k_y(A_2+A_4-A_5)k_y + k_z(A_1+A_3)k_z + (D_2+D_4-D_5)(e_{xx}+e_{yy}) + (D_1+D_3)e_{zz}, \quad (2)$$

where e_{ij} is the strain tensor, $a_c^{\parallel}$ and $a_c^{\perp}$ are the conduction-band deformation potentials, A_i $(i=1,\ldots,5)$ are the Rashba-Sheka-Pikus valence-band parameters, D_i $(i=1,\ldots,5)$ are valence-band deformation potentials, $U_{c(v)}$ is the energy of the unstrained conduction- (valence-)band edge. The electron (hole) projection of exciton ground-state wave function $\Psi_{e(h)}$ is obtained by solving the Schrödinger equation self-consistently,

$$(H_{e(h)} + V_{h(e)})\Psi_{e(h)} = E_{e(h)}\Psi_{e(h)}, \quad (3)$$

$$\epsilon_0 \nabla(\epsilon_s \nabla V_{h(e)}) = \pm e|\Psi_{h(e)}|^2. \quad (4)$$

Exciton emission energy is written as

$$E_{ex} = E_e - E_h + (\langle\Psi_h|V_e|\Psi_h\rangle - \langle\Psi_e|V_h|\Psi_e\rangle)/2. \quad (5)$$

All the material parameters are taken from Ref. 20 except the value of spontaneous polarization. As we shall discuss later, the small spontaneous polarization difference between GaN and AlN or the small built-in electric field is consistent with the experimental results.

Figure 2(b) shows the calculated electron and the hole wave functions. In the absence of the external electric field, the hole wave function is localized near the bottom of the dot, while the electron wave function is localized near the top due to the piezoelectric field and the pyroelectric field. In the presence of an external lateral electric field, the hole wave function is shifted horizontally to the direction of the applied field, while the electron wave function remains confined near the center of the dot top. The difference between the electron and the hole arises from the different effective mass and lateral confinements.

Calculated field dependence of the electron (E_c) and the hole confined state energies (E_v) are shown in the inset of Fig. 3(a). With increasing field, E_c decreases and E_v increases in a high field range above 200 kV/cm. These are conventional field dependences caused by the tilting of their band edges. On the other hand, in a low field range less than 150 kV/cm, E_c increases with increasing field. This unconventional increase of E_c is a consequence of a sharp bending of the confining potential of the electron near the horizontal dot edge. Along the x direction, the potential rapidly increases by about 0.1 eV at the horizontal dot edge. The bowl-shaped potential profile is sharper than parabolic. Such a bending of the potential has also been seen in another calculation.[21] In our size and shape of dots, the bending potential increases the energy level of the confined electron state with increasing the field. On the other hand, the hole confined state energy (E_v) shows a conventional monotonic

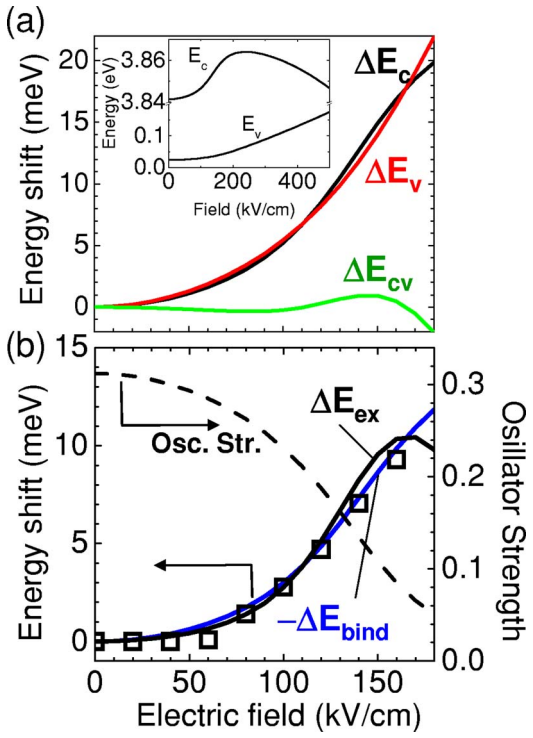

FIG. 3. (Color online) (a) Calculated energy shifts of electron energy level (ΔE_c), hole energy level (ΔE_v), and transition energy without excitonic effect [$\Delta E_{cv}=\Delta(E_c-E_v)$] as a function of the electric field. (b) Calculated variation of the binding energy ($-\Delta E_{\text{bind}}$) and the Stark shift of the excitonic state [$\Delta E_{ex}=\Delta(E_c-E_v-E_{\text{bind}})$]. The experimentally measured energy shift (squares) averaged for positive and negative fields is also shown. The dashed line represents the oscillator strength.

increase with the field because the hole state has a much heavier effective mass than the electron. The hole state is less affected by such a peculiar potential due to much weaker confinement or stronger localization.

In the low field range, the ground transition energy without Coulomb interaction $E_{cv}=E_c-E_v$ becomes roughly independent of the field. With Coulomb interaction the exciton emission energy decreases by the binding energy. The exciton binding energy, which is 31.1 meV at zero field, rapidly decreases with an increasing field mainly due to the large movement of the hole wave function [Fig. 2(b)]. At 150 kV/cm, the binding energy is reduced by 10 meV. Thus, the shift of the exciton emission energy ΔE_{ex} is dominated by the decrease of the exciton binding energy $|\Delta E_{\text{bind}}|$ [Fig. 3(b)]. The resultant blueshift of the emission energy agrees well with the experimental one. With further increasing the electric field, the effect of the peculiar confining potential is compensated by the band-tilting effect, and consequently, the exciton emission energy starts to decrease. The decrease of the emission energy has not been observed in our experiment because the emission intensity rapidly decreases due to large spatial separation of the electron and hole. The decrease of the intensity is consistent with the calculated decrease of the oscillator strength [Fig. 3(b)].

The unconventional increase of emission energy is influenced by the magnitude of the built-in internal electric field.

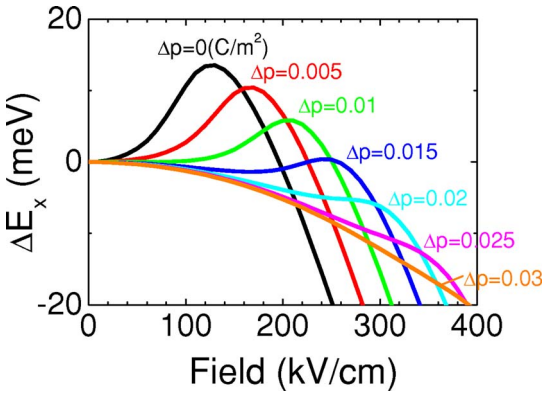

FIG. 4. (Color online) Calculated energy shifts of the Stark shift of the excitonic state plotted for various spontaneous polarization differences Δp.

Generally, the reported magnitude of the built-in field in the GaN/AlN system strongly depends on the sample or the growth condition. Although several reasons for the diverse magnitude have been invoked,[22,24] the reason has still been controversial, to our knowledge. For simplicity, we only vary the spontaneous polarization difference between GaN and AlN, although we do not rule out the possibility of different piezoelectric constants or strain. In our calculation, the zero polarization difference corresponds to a built-in field of 1.2 MV/cm at the center of the dot, while the polarization difference $\Delta p=0.052$ in Ref. 20 obtained by a first-principle calculation[26] corresponds to 4.3 MV/cm. The model dot which reproduces the experimental Stark shift [Fig. 3(b)] has a much smaller spontaneous polarization difference of $\Delta p=0.005$ C/m^2 than the value of $\Delta p=0.052$ C/m^2 in Refs. 20 and 26, as shown in Fig. 4. Such a small spontaneous polarization difference has been reported in several experiments.[27,28] A reduction of the spontaneous polarization might be caused by segregation of the species at the dot boundaries during growth, yielding nonplanar interfaces.[25] The small spontaneous polarization difference or the small built-in field is appropriate also to match the radiative lifetime. The calculated lifetime of 2 ns agrees with the experimental lifetime of our dots[29] of about 3 ns at the emission energy, while the calculation using the conventional spontaneous polarization ($\Delta P=0.052$ C/m^2) results in a lifetime of 40 ns.

The model dot whose energy shift agrees with the experiment has a slightly larger height-to-base aspect ratio (4 nm:16 nm) than the typical ratio obtained from the TEM measurement (3 nm:20 nm), although the difference is within the inhomogeneous dot-size distribution in our sample. The calculations for the GaN/AlN dots with different sizes and aspect ratios show that the unconventional behavior persists until the dot height is larger than or equal to 4 nm and the base diameter is smaller than or equal to 22 nm. In the dots with small height-to-base aspect ratios, the large top diameter makes the electron less affected by the peculiar confining potential near the dot edge. In addition, large top and base diameters increase the band-tilting effect of the conduction- and valence-band edges, leading to a

conventional Stark shift.

In conclusion, the measured nearly symmetric blueshift in the self-assembled GaN dots is attributed to a strong excitonic effect and a peculiar confining potential. The unconventional Stark shift allows a direct measurement of the exciton binding energy tuned by up to 10 meV. The tunability of the electrical dipoles will be important in the applications for GaN-based single-photon sources and for future quantum information technology.

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