Measurement of an Exciton Rabi Rotation in a Single GaN/$Al_xGa_{1-x}N$ Nanowire-Quantum Dot Using Photoluminescence Spectroscopy: Evidence for Coherent Control

M. Holmes,[1,*] S. Kako,[1,2] K. Choi,[2] P. Podemski,[1] M. Arita,[1] and Y. Arakawa[1,2]

[1]*Institute for Nano Quantum Information Electronics, The University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo 153-8505, Japan*
[2]*Institute of Industrial Science, The University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo 153-8505, Japan*

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Experimental observation of excited state exciton Rabi rotation in a single GaN quantum dot is presented. The dot is embedded in a site-controlled GaN/AlGaN nanowire. Damped oscillation is observed in the power-dependent spectra of the quantum-dot ground state upon resonant pumping of an excited state that had been identified by photoluminescence excitation spectroscopy. A discussion on the origins of the damping is given.

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Coherently manipulating qubits is an integral process to the successful execution of quantum-information processing (QIP). Although the realization of QIP in the solid state is a challenge fraught with difficulties, the optical excitation and manipulation of excitonic or spin states in quantum dots (QDs) is considered to be one promising avenue of investigation [1–3]. Indeed, several techniques have been adapted to perform the coherent optical control of excitonic qubits in QDs by addressing them with pulsed lasers.

In the case of a qubit formed by a pure two-state system ($|\Psi\rangle = \alpha|0\rangle + \beta|1\rangle$), the applied optical field acts to coherently rotate the state vector back and forth between the constituent states, resulting in an observable oscillation (Rabi rotation) in some choice of output measurement sensitive to the state populations. If the system is initially in state $|0\rangle$, and with no dephasing, the postpulse population of state $|1\rangle$ oscillates as $\sin^2(\Theta/2)$, where the excitation pulse area Θ is defined as $(1/\hbar)\int_{-\infty}^{\infty} \mu\epsilon(t)dt$, μ is the transition dipole moment, and $\epsilon(t)$ is the laser pulse field polarization vector. The observation of this rotation is a clear sign of the successful coherent manipulation of the qubit. For $\Theta = \pi$ the population of the upper state is maximized. Such an optical π pulse is the QIP equivalent of a NOT logic gate (the same pulse applied to a system in state $|1\rangle$ will invert the state vector back to $|0\rangle$).

To date, the majority of quantum-dot coherent control experiments have been performed on QDs formed in the III-As semiconductor system, with which a two-qubit controlled rotation (CROT) logic gate has also been realized using exciton and biexciton states [4]. However, there have been no reports of the successful coherent optical manipulation of III-nitride QDs (which emit in the UV to visible regions). The III-nitride system is promising as it can sustain room-temperature stable excitons, a property which enabled the realization of a single photon emitter operating at 200 K [5]. It has also been theorized that coupled GaN wurtzite QDs could be good candidates for the implementation of ultrafast QIP logic operations [6]. In this Letter, we present experimental evidence of the observation of power-dependent Rabi rotation in a GaN quantum dot. We control a qubit defined by the crystal ground state $|0\rangle$ and an excited excitonic state $|1\rangle$ in the dot, using the excitonic ground state $|s\rangle$ emission to measure the oscillation.

The GaN QDs investigated in this study are formed near the tips of site-controlled GaN/$Al_xGa_{1-x}N$ nanowires of nominal mole fraction $x = 0.8$. A SEM image of a single nanowire, along with a schematic depiction of the structure can be found in Fig. 1. The wires themselves were grown by metal-organic chemical vapor deposition, at sites defined by apertures opened in a SiO_2 mask, and consist of a GaN core section surrounded by the $Al_xGa_{1-x}N$ dot barrier. The GaN QD was formed by a further GaN growth step before being capped with $Al_xGa_{1-x}N$. Growth conditions can be found elsewhere [7,8]. The quantum dots are expected to take the form of hexagonal disks with a lateral dimension of ~10 nm and a height of ~1 nm. The mask pattern for selective growth had a 20 μm pitch, resulting in spatially separated devices which could be probed individually.

The QDs were cooled to 3.8 K in a continuous-flow helium cryostat and addressed at a steep angle (~ 60° to the nanowire axis) with a frequency-tripled tunable Ti:Al_2O_3 laser (at 80 MHz, 200 fs pulses). The excitation

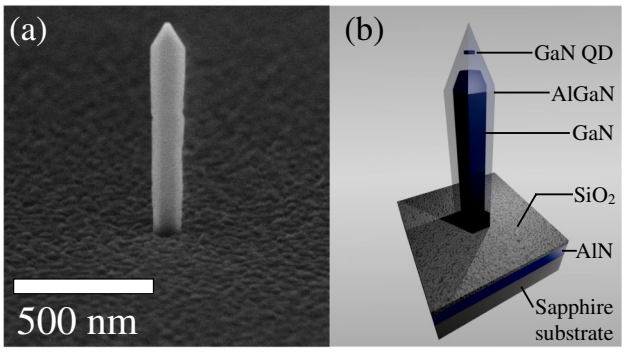

FIG. 1 (color online). (a) SEM image of a single GaN/$Al_xGa_{1-x}N$ nanowire containing a GaN quantum dot. (b) Schematic view of the nanowire-QD structure.

laser beam width has been measured to be ~7 meV (FWHM). The photoluminescence from the sample was collected normal to the sample surface by a separate 50× objective (numerical aperture 0.4) and directed to a grating spectrometer (1200 lines/mm and blaze 300 nm) before being measured with a CCD array. The spectral resolution of the setup was ~2 meV. A tunable $4f$ spatial filter was used to isolate the QD signal from the rest of the light collected by the objective. A schematic of the experimental setup can be found in Fig. 2.

The nanowire QDs are measured to emit typically at energies in the range of 4.2–4.5 eV upon excitation at 4.65 eV (266 nm), and the measured linewidth of the dot emission is typically resolution limited. This linewidth is typical for GaN QDs [9], and is most likely caused by spectral diffusion induced inhomogeneous broadening [9,10]. The emission energy indicates that the QDs are indeed small, and this may be a crucial factor for the successful coherent control, as is discussed below.

In order to probe the excitonic excited states of a single quantum dot, photoluminescence excitation (PLE) spectroscopy was performed by tuning the excitation laser energy while measuring the ground state emission intensity from a single quantum dot. The nanowire was continuously monitored during the tuning process, and any displacement of the excitation beam was automatically compensated for by two piezo-mounted mirrors and beam profilers. Additionally, any sample movement was tracked under white light illumination, and corrected for, between data acquisitions by the automatic translation of cryostat stage. The stability afforded by this active beam position stabilization system allowed for the successful excitation of a single dot while tuning the frequency-tripled light source [11].

The photoluminescence spectrum along with the corresponding PLE spectrum of a single GaN QD embedded in a nanowire is presented in Fig. 3(a). The single peak in the PL spectrum at 4.375 eV is assigned as the ground state emission of the QD ($|s\rangle \rightarrow |0\rangle$). The PLE spectrum is comprised of a clear absorption peak at ~4.48 eV and a rising background that increases with excitation photon energy. The background is attributed to absorption into (and the subsequent relaxation from) a continuum of states [12–15], whereas the prominent peak is most probably due to transitions between one or more excited excitonic states and the ground state of the QD (absorption into the excited state, emission at the ground state). Indeed, this PLE peak appears to have one resolution limited component (limited by the excitation laser linewidth), and a broader shoulder on the high-energy side. Eight-band $k \cdot p$ calculations for hexagonal disk-shaped $GaN/Al_{0.8}Ga_{0.2}N$ quantum dots support the identification of the peak in the PLE spectrum as originating from direct excitation of an excited state [see Figs. 3(b) and 3(c)], and tentatively indicate a QD size of approximately 0.85 × 7 nm. Probing the excitonic structure of the dot in this way enables the identification of candidate states for resonant excitation during the coherent control process. Successful coherent control requires the excitation of a single transition in the dot, and also that the interaction time be shorter than the total coherence time of the system.

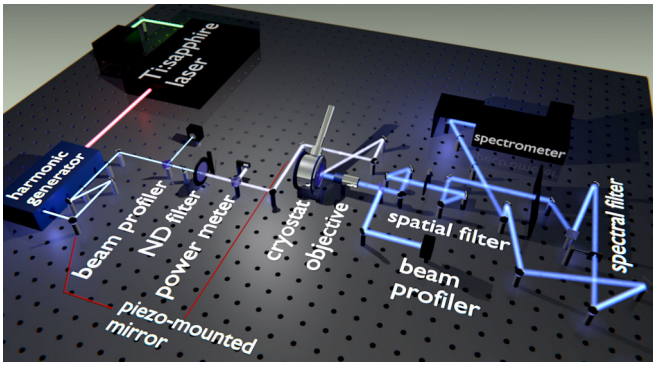

FIG. 2 (color online). Schematic of the experimental microphotoluminescence-PLE system.

The resolution limit of the PLE experiment prevents the acquisition of any knowledge on the excited state dephasing time and, furthermore, the inhomogeneously broadened ground state emission linewidth also denies the possibility of making any meaningful estimate on a lower bound for the dephasing time. We do know, however, that the lifetime of the ground states from these dots are typically measured to be ~250 ps using time resolved photoluminescence techniques (not shown here). The excited state lifetime should be faster than this.

Previous theoretical investigations on the energy structure of GaN quantum dots show that smaller dots have a

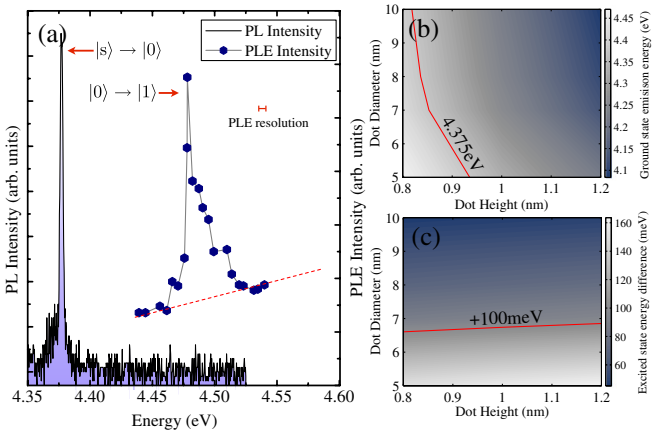

FIG. 3 (color online). (a) PL and PLE spectra of the quantum dot under investigation (measured at $T = 3.8$ K). Panels (b) and (c) show energy maps of the exciton ground and excited state energies calculated for hexagonal disk shaped $GaN/Al_{0.8}Ga_{0.2}N$ QDs using 8 band $k \cdot p$ theory including the effects of strain and exciton binding. The red contour lines indicate the QD dimensions that have the excitonic energy levels measured experimentally. These calculations tentatively indicate a QD size of approximately 0.85 nm × 7 nm, which is in good agreement with our expectations.

greater energy separation between states [16–18], and Winkelnkemper *et al.* [16] have shown that some optically active *p*-shell states can be separated by as much as 20–25 meV (for a slight 10% anisotropy in the lateral dimension of the dot). In addition, Tomić and Vukmirović have shown a similar state splitting when including the excitonic spin orbit interaction [18], and Kindel *et al.* have calculated that we may expect to observe fine structure splitting of order 12 meV [19]. Furthermore, other studies have shown an increased biexciton binding energy (20–40 meV) in small nanowire quantum dots [20]. It therefore seems reasonable that for the small QDs under investigation here, we could expect to observe energy separations of over 10 meV between individual excitonic states (depending heavily on the individual dot geometry). Therefore, by using these small QDs it should be possible to address and control a single GaN QD excitonic state with an ultrafast laser pulse. We here assume that the sharp component of the peak in the PLE spectrum of Fig. 3 is due to the excitation of a single excitonic state, $|1\rangle$, and having identified the required states, can attempt the coherent control.

In order to experimentally address the qubit defined by states $|0\rangle$ and $|1\rangle$, the laser was tuned into resonance with the $|0\rangle \leftrightarrow |1\rangle$ transition, and the intensity of the ground state emission was monitored while the excitation power was controlled. The dependence of the integrated ground state recombination PL signal on the excitation power on is shown in Fig. 4(a). The figure inset depicts a simplified QD electronic structure including the relevant states and transitions. State $|s\rangle$ is populated by nonradiative decay from $|1\rangle$ (the process which allows the PLE measurement to be made). The actual PL spectra at various excitation densities (for both resonant- and nonresonant excitation conditions) are shown in Figs. 4(b) and 4(c), respectively. Damped Rabi rotations are observed in the ground state PL intensity

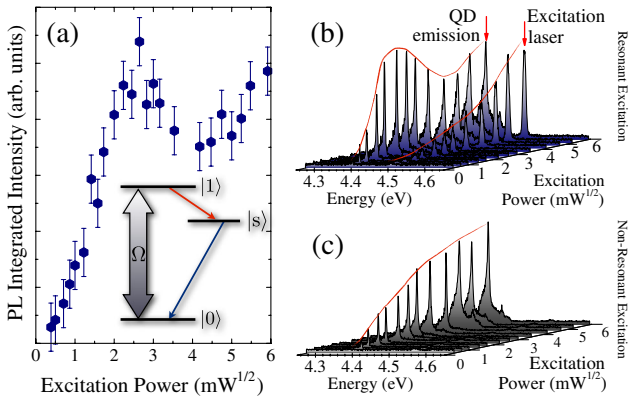

FIG. 4 (color online). (a) Power-dependant Rabi oscillations observed in the excitonic ground state emission upon resonant excitation of the excited state. The inset shows a simplified energy level diagram of the system. Panels (b) and (c) show the emission spectra for the resonant and nonresonant excitation cases, respectively. The red lines are a guide to the eye.

as the pulse area (proportional to the square root of the excitation power) is increased. In the case of nonresonant excitation, the expected monotonic increase in emission intensity is recovered, although with an increased intensity at higher excitation powers, most likely fueled by further repopulation of the dot from the continuum states.

The damping of the observed oscillation may be due to a complex interplay between several processes involving time-averaging and dephasing effects. Here we give a brief consideration to some of these processes, but note that it is difficult to be definitive in our analysis, due to the measurement limit of the experimental data. First, we believe that the continuum states play a role in the observed damping. This could be by both the incoherent population of the dot via relaxation from the continuum states [13,15,21], and also due to an interaction between the addressed qubit and fluctuations in the continuum. These interactions will result in temporal fluctuations of the optical dipole, which when averaged (during the acquisition of the emission spectrum) can lead to an observed intensity damping. Similarly, time varying interactions with charge-populated defects in the vicinity of the dot (which give rise to spectral diffusion [9,10,22]) can also be expected to induce dipole fluctuations, and hence damping. We do note, however, that the degree of spectral diffusion measured in these dots is small, and as such this effect may also be small (though it can be expected to increase at higher excitation densities [10]).

Another major cause of the damping may be the excitation of more than one state in the dot, the likelihood of which is increased by the lack of polarization control in our current setup. As mentioned above, the energy separation of excited states within a shell could be several tens of meV depending on the geometry of the exact dot under investigation. While this is larger than our resolution limit, we do observe a shoulder to the peak in the PLE spectrum, which could be due to a second state. The excitation of different states (with different optical dipoles) will not necessarily preclude the observation of oscillation [23], but will lead to an observed decay. Furthermore, a further excitation to a biexciton state will result in a breakdown of the $|0\rangle \leftrightarrow |1\rangle$ two level system and a decay of the oscillation.

The interaction with acoustic phonons at the Rabi energy will lead to exciton dephasing, resulting in a power-dependent damping of the Rabi oscillations [24–26]. This may be a cause of the damping observed in our experiment, although an analysis cannot be performed at present due to the fact that we only observe two turning points (at least three points are required to discern the degree of power damping).

Phonons may also act to reduce the (presently unknown) lifetime of $|1\rangle$ via a phonon-mediated $|1\rangle \rightarrow |s\rangle$ relaxation process. The phonon population will increase at elevated temperatures, so it is possible that these effects will become more important at higher excitation powers due to sample heating. Indeed, we do observe an asymmetric

broadening of the ground state emission peak at increased excitation power (consistent with acoustic phonon coupling [27]), and furthermore measure a small ground state peak emission shift of ~ 600 μeV as the pulse power is increased from a π pulse to a 2π pulse. This shift is most probably due to band gap shrinkage upon sample heating [28] but is over an order of magnitude smaller than the spectral width of the excitation laser pulse, and should not directly effect the observation of the Rabi oscillations.

Finally, while we are limited to a qualitative discussion on the origins of the damping here, we finish by noting that the observation of Rabi oscillations indicates that the dephasing time of the system is larger than the interaction time with the laser, i.e., >200 fs. Further investigation will be required to fully understand the dynamics of the system in question.

In conclusion, the coherent control of a site-controlled GaN quantum dot in a GaN/AlGaN nanowire has been successfully achieved. At this stage, it is believed that the relatively large energy separation of excited states due to the small size of the QD enabled the excitation of a single excited state. This is a tentative step toward the realization of quantum-information processing with III-nitride systems, a material system that may allow us to harness solid state QIP at room temperature. The exact cause of dephasing is not currently known, and requires further investigation. We envisage that with the addition of polarization control, and pulse shaping to reduce the excitation pulse width, we may be able to probe the excited states in more detail, obtain higher-quality Rabi oscillations, and hence study these dephasing mechanisms. In the near future it may be possible to generate single photons on demand from the ground state by resonant excitation of the excited state with a π pulse [29]. The use of site-controlled QDs is of further benefit as this may facilitate the future development of arrays of devices.

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*holmes@iis.u-tokyo.ac.jp

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