Nanosecond-scale spectral diffusion in the single photon emission of a GaN quantum dot

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Nanosecond-scale spectral diffusion in the single photon emission of a GaN quantum dot

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Autocorrelation measurements are used to reveal the spectral diffusion time scale in the single photon emission of a GaN interface fluctuation quantum dot. Typical characteristic diffusion times of such QDs are revealed to be of nanosecond order. The excitation power dependence of the diffusion rate is also investigated, whereby an increase in the diffusion rate with increasing excitation power is observed. This result provides information on experimental conditions that will be required for the generation of indistinguishable photons. https://doi.org/10.1063/1.4997117

Spectral diffusion is an emission linewidth limiting phenomenon in quantum dots (QDs) involving random jumps in the emission wavelength. It usually occurs from electronic interactions between a confined exciton and its environment (such as charge fluctuations).[1–4] Such energy fluctuations will ultimately limit the indistinguishability of the emitted photons, and therefore have detrimental effects on the use of single photon emitters in future quantum applications.[5–10] Spectral diffusion in quantum dots is therefore an important topic of research interest, and has been the subject of several recent theoretical and experimental studies. Indeed, great efforts have been made to understand and suppress spectral diffusion in QDs of various material systems,[11–15] particularly those that emit in the visible and infra-red. Recently, through suppression of the formation of charge traps, linewidths as low as 30μeV have been obtained from InAsP quantum dots in inP nanowires.[16,17]

As a subset of semiconductor quantum dots, III-Nitride based quantum dots have attracted extensive attention due to their wide emission wavelength range from the ultraviolet to the infrared[18–21] and their possible high-temperature operation.[22–26] However, typical III-Nitrides suffer from a large degree of spectral diffusion, which can broaden the emission linewidth up to the order of a few meV.[1,2,27] Typically, spectral diffusion on such large energy scales is investigated by measurement of the temporal variation of the luminescence on long-time scales. Indeed, this method has been used to evaluate temporal fluctuations in Nitride based QDs,[28,29] and also other materials such as CdTe QDs,[13] CdSe QDs,[14] and InAlAs QDs.[15]

However, fluctuations can occur on timescales varying from sub-nanosecond[30] levels to several seconds,[28] and measuring the fluctuations on fast time scales requires more advanced measurement techniques such as photon autocorrelation.[30] This converts the spectral fluctuation into an intensity fluctuation and therefore benefits from the sub-nanosecond time resolution of photomultiplier tubes (PMTs) used in a Hanbury Brown & Twiss type setup. Although spectral diffusion in different materials has been investigated using this method, including CdSe QDs embedded in ZnSe nanowires,[30–32] nitrogen vacancy centers in nano size diamonds[33] and oxidized tungsten disulfide multilayers,[34] the fast spectral diffusion in III-Nitride QDs has yet to be investigated.

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In this letter we elucidate the spectral diffusion timescale of a single-photon-emitting GaN interface fluctuation QD[35] via photon autocorrelation measurements, and furthermore provide information on the characteristic spectral diffusion time as a function of excitation laser power. The studied interface fluctuation GaN QD sample was grown by metalorganic chemical vapor deposition (MOCVD), and particular growth details can be found elsewhere.[35] For measurement, the sample was mounted in a continuous flow helium cryostat with a cryogenic temperature controller (Oxford Instruments ITC503), cooled to 10K, and excited with a 266nm wavelength continuous wave diode pumped solid-state laser (CryLaS FQCW 266).

Micro-photoluminescence from the sample was collected using a UV objective lens (40x, NA=0.6), and detected with a nitrogen cooled CCD array attached to a 75cm spectrometer with a 2400mm^{-1} diffraction grating. The spectral resolution of the setup was ~200μeV. Autocorrelation of the emission was performed by using a standard UV HBT setup with two photomultiplier tubes (Hamamatsu H10682-210) combined at the exit slit of the spectrometer. The time resolution of our setup has been measured to be ~400ps.

We begin by investigating the temporal evolution of the emission spectrum of an excitonic emission line from a single QD excited at a laser power of 200μW, measured over a 1-hour period (each spectrum was integrated for 5s). As can be seen in figure 1, the emission is extremely stable. Indeed, the left inset of the figure shows the time evolution of the peak center, where it is clear that the extent of any spectral wandering (long term spectral diffusion) is much smaller than the energy resolution limit of our system.

However, as shown in the right inset (selected spectra at several different times), the lineshape of the emission has a fairly static Gaussian profile, indicating that the emission energy is undergoing some stochastic fluctuations on time scales much faster than the shutter speed of our CCD. In order to further investigate the properties of this fast spectral diffusion, we resort to a higher resolution measurement using the autocorrelation of the emission.

Figure 2a shows the same emission spectrum of the GaN interface fluctuation QD integrated over 1s. The full width at half maximum (FWHM) of the emission peak of this particular dot is measured to be ~300 μeV. By changing the angle of the grating in the spectrometer and the size of the exit slit connecting the spectrometer and HBT setup, we could freely select a spectral window for autocorrelation measurement.

Two dashed boxes in figure 2a represent different spectral regions that were selected for the measurements shown in figures 2b and 2c. Figure 2b corresponds to a measurement of the whole peak (grey dashed box in figure 2a), which represents a ~700 μeV spectral window width. The data in figure 2c corresponds to the photon autocorrelation measurement of one half of the emission peak (the blue dashed box in figure 2a) with a spectral range of ~300 μeV.

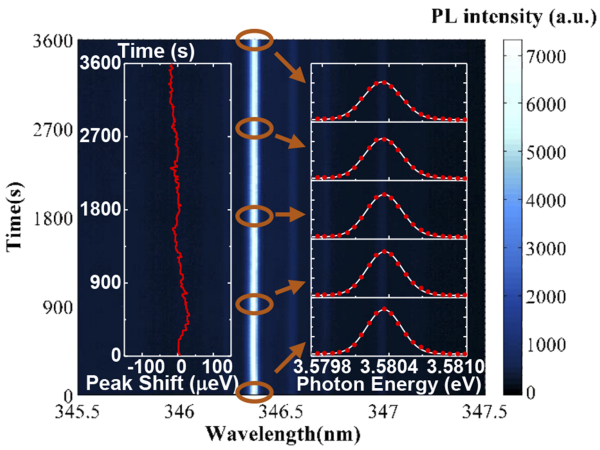

FIG. 1. Temporal variation of the emission spectrum of a selected single GaN interface fluctuation quantum dot measured over a period of one hour. The left inset shows the peak shift of the emission, and the right inset shows five representative spectra (data points) with Gaussian lineshape fits (white lines).

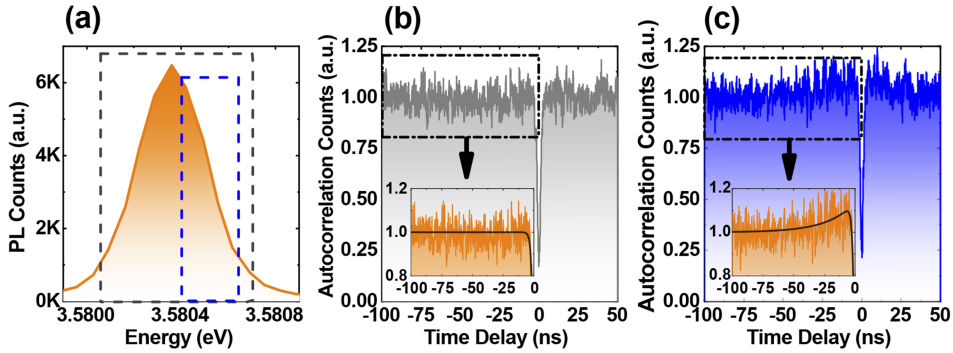

FIG. 2. Spectral diffusion measurements using autocorrelation. (a) Photoluminescence spectrum of a single GaN interface fluctuation QD. (b) Autocorrelation of the whole peak as the grey dash box in figure 2a. (c) Autocorrelation of high energy profile range as the blue dash box in figure 2a. The inset figures in (b) and (c) show a zoomed in region of the autocorrelation measurement such that the bunching effect can be clearly seen.

When the full peak is measured (figure 2b) a typical narrow antibunching ($g^{(2)}(0)<0.5$) in the photon statistics clearly reveals the single photon nature of the emission, as expected for a quantum two level system. However, when only one half of the emission peak is measured (figure 2c), the spectral diffusion of the peak into and out of the measurement window induces an additional photon bunching in the autocorrelation (in addition to the clear single photon emission).

In order to evaluate the bunching time scale, the data has been fitted with a second order correlation function including a bunching envelope to describe the spectral diffusion:[30]

$$g^{(2)}(\tau) = [1\text{-}\alpha\exp(\frac{-|\tau|}{\tau_e})]^*[1 + \beta\exp(\frac{-|\tau|}{\tau_{SD}})],$$

where the factors α and β represent the extent of the antibunching dip and the visibility of the bunching peak, respectively. Here τ is the time delay of the autocorrelation measurement, τ_e is the time scale of the antibunching dip (in this case ~1ns), and τ_{SD} is the time scale of the spectral diffusion that we focus on here. We find that the characteristic spectral diffusion timescale of this dot under these conditions is ~22ns. We note that this value is comparable with previous reports of spectral diffusion in self-assembled CdSe QDs.[30]

As spectral diffusion is induced by the fluctuating Stark shifts[36] due to the charge environment, it is expected to exhibit an excitation power dependence whereby the diffusion rate increases with increased number of excited charges in the environment. In order to investigate the relation between the excitation power and the spectral diffusion of the fluctuation GaN QD, we varied the excitation power of the laser from 100μW to 500μW and performed the autocorrelation measurement as shown in figure 3a. The spectral diffusion characteristic rate (the inverse of the spectral diffusion characteristic time) has been extracted as above, and is shown as a function of excitation power in figure 3b. The figure inset also shows the emission linewidth of the GaN QD under various powers, revealing that it only changes by a small amount as the excitation power is increased. However, the measurement results show that the diffusion rate increases with increasing excitation power.

Such an increase of the spectral diffusion rate with increasing excitation power has been observed in other material systems.[31,33,34] In previous works, this relationship between the excitation power and the spectral diffusion rate has been observed with a linear dependence in diamond-based Nitrogen vacancy centers[33] and with a sublinear dependence in CdSe QDs embedded in ZnSe nanowires.[31] However, in the present case, we do not observe any apparent saturation in the diffusion rate with increasing power over the studied excitation power range. The observed relationship between the excitation power and the diffusion rate can be approximated with a linear trend as shown by the black arrow in figure 3b. It is also apparent the diffusion rate is directly proportional to the excitation power, in that the rate tends towards zero at zero excitation power- i.e. in the limit of no excited carriers.

For single photon sources, this spectral diffusion phenomenon will affect the indistinguishability of the photon emission. Our results show here that it may be possible to generate indistinguishable photons using III-Nitride QDs under excitation with pulse pairs separated by times less than ~10ns.

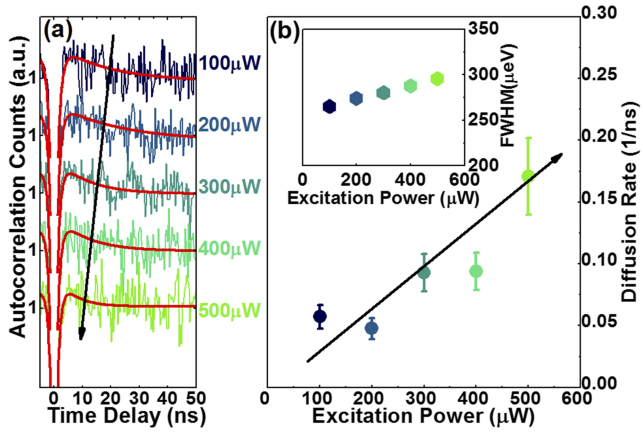

FIG. 3. (a) Power dependent photon autocorrelation data (log scale), offset for clarity. (b) Excitation power dependence of the spectral diffusion rate. The black arrows are guides to the eye. The inset in (b) shows the change of the emission linewidth under various power.

In summary, we have reported a study of spectral diffusion in a GaN QD and revealed the characteristic spectral diffusion time, via measurement of the second order autocorrelation, to be of order ~20ns. We have also presented the excitation power dependence of the spectral diffusion rate, showing the expected increase in rate as the excitation power is increased. Such information on the spectral diffusion times will be important for the generation of indistinguishable photons using this material system.

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