Memorandum Vol. 5, No. 8 / August 2018 / Optica 932

Resonant excitation of quantum emitters in gallium nitride

Mehran Kianinia,[1] Carlo Bradac,[1] Minh Nguyen,[1] Tongtong Zhu,[2] Milos Toth,[1] Rachel Oliver,[2] and Igor Aharonovich[1,*]

[1]Faculty of Science, University of Technology Sydney, Ultimo, New South Wales 2007, Australia
[2]Department of Materials Science and Metallurgy, University of Cambridge, Cambridge CB2 3QZ, UK
*Corresponding author: igor.aharonovich@uts.edu.au

Received 5 July 2018; accepted 11 July 2018 (Doc. ID 337872); published 30 July 2018

We demonstrate resonant excitation of quantum emitters in gallium nitride (GaN). The emitters are stable under non-resonant excitation and exhibit nearly Fourier-transform-limited lines of ~250 MHz under coherent excitation, the narrowest reported to date for GaN.

https://doi.org/10.1364/OPTICA.5.000932

Advance quantum nanophotonics technologies rely on the ability to resonantly excite quantum emitters to achieve Fourier transform (FT)-limited linewidths [1–3]. Among various solid-state quantum systems, point defects in diamond [4,5], SiC [6,7], and hexagonal boron nitride [8,9], as well as carbon nanotubes [10], have attracted the most attention due to their exceptional brightness and photostability. Recently, quantum emitters in gallium nitride (GaN) have emerged as promising candidates for integrated quantum photonics, due to the presence of quantum emitters in the ultraviolet [11], visible [12], and the telecom [13] spectral ranges, and the availability of mature growth and fabrication protocols for this material.

Despite progress in room temperature characterization of defects in GaN, coherent (or resonant) excitation properties of the emitters in GaN have not been reported to date. Coherent excitation provides essential information about the emission linewidths and photophysical properties of the emitters. It is the most crucial step toward the generation of indistinguishable photons, entanglement, and realization of coherent control of the emitters' quantum states.

Here we study the optical properties of quantum emitters in GaN at cryogenic temperatures under both coherent and incoherent excitation. The emissions have a full width at half-maximum (FWHM) of ~2 nm under non-resonant excitation and exhibit stable emission. Resonant excitation yields nearly FT-limited lines with a FWHM of ~250 MHz.

In our experiments, we used a 4.2 μm thick low-defect-density layer of hetero-epitaxial GaN grown with no intentional doping by metal-organic vapor phase epitaxy on a c-plane sapphire substrate using the method described in [14], as shown in the inset in Fig. 1(a). Here we focus on emitters in the visible spectral range. The emitters were excited using a 707 nm titanium sapphire (Ti:SAP) laser. In the detection path, the laser was rejected with a 715 nm long-pass filter and emissions in the spectral range of 720–900 nm were collected. All measurements in this work were performed at 10 K. A high-resolution photoluminescence (PL) spectrum of an emitter in GaN is shown in Fig. 1(a). It exhibits a sharp zero phonon line (ZPL) at 746 nm with a FWHM of 2 nm.

To check the emitter's stability under the same excitation conditions, we measured successive PL spectra over 90 s using a 1 s acquisition time. As shown in Fig. 1(b), spectral diffusion is very subtle, and the PL is stable within the time resolution of the measurement. This is highly advantageous for scalable applications of quantum emitters, where photostability is considered a prime factor for practical devices.

Next, we performed coherent excitation of the emitter to gain insights into the natural linewidth and broadening mechanisms of the ZPL. The average lifetime of emitters in GaN has been reported to be 2.5 ns [12], yielding an FT-limited linewidth of ~65 MHz. For this particular emitter, we measured a lifetime of ~1.1 ns, which corresponds to ~150 MHz.

The same continuous-wave Ti:SAP laser (50 kHz linewidth; MSquare) was swept over 5 GHz using a step size of 20 MHz to coherently excite the emitter shown in Fig. 1. The excitation laser was filtered with a 750 nm long-pass filter and the PL emission in the phonon sideband was collected. A resonant excitation spectrum recorded at 5 μW is shown in Fig. 2(a). Experimental data (red dots) were fitted with both Lorentzian (violet) and Gaussian (orange) functions with linewidths of 219 and 310 MHz, respectively. The Gaussian function yields the better fit, with χ^2 of 230, versus 305 for the Lorentzian fit. This indicates that broadening of the ZPL is likely caused by spectral diffusion rather than homogeneous broadening mechanisms such as interaction with lattice phonons [15]. Note that the room temperature spectrum is broadened due to thermally activated phonons.

The effect of spectral diffusion on emission under coherent excitation is shown in Fig. 2(b). For this measurement, the laser was tuned to the optical transition energy of the emitter, and the PL was acquired for 10 min. Note that in Fig. 2(b), the PL

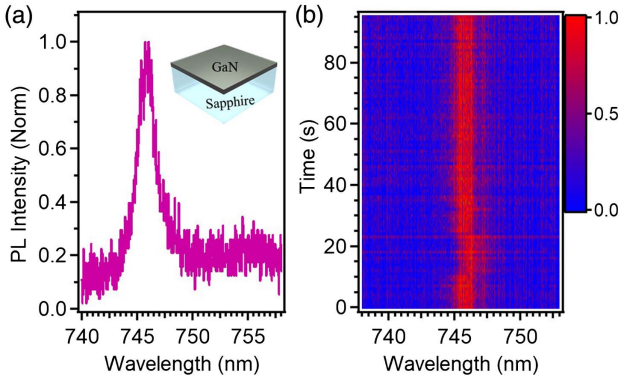

Fig. 1. PL of emitters under non-resonant excitation. (a) A high-resolution spectrum of an emitter excited using a 300 μW, 707 nm laser. The inset shows the structure of the GaN sample: 4.2 μm of c-plane GaN grown on a sapphire substrate. (b) Successive PL spectra of the same emitter collected every second, demonstrating stable emission with negligible spectral diffusion.

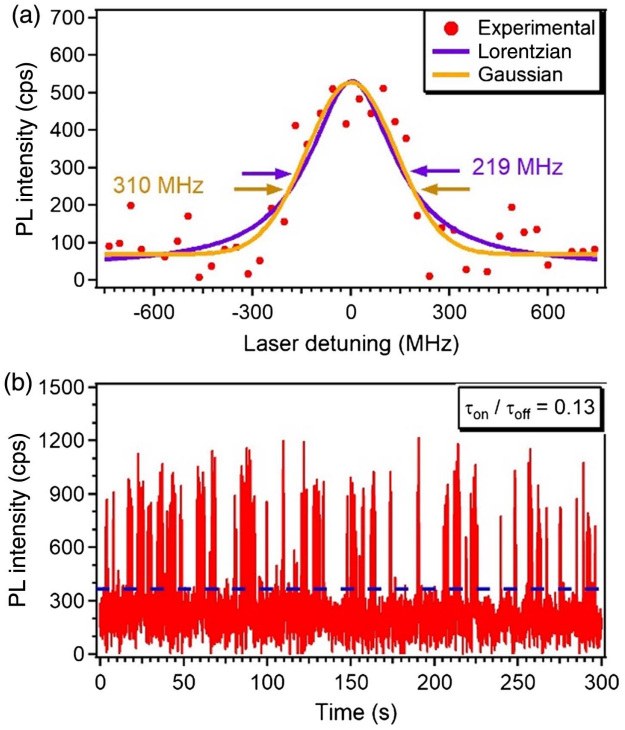

Fig. 2. PL under coherent excitation. (a) Resonant PL excitation measurement of the emitter in Fig. 1. The experimental data have been fitted with Gaussian (orange) and Lorentzian (violet) functions with FWHMs of 310 and 219 MHz, respectively. (b) PL emission of the emitter under fixed coherent excitation. The dashed line is the threshold used for calculating the on–off ratio of 0.13.

intensity trace is shown for 5 min to clearly illustrate the intermittency (blinking) of the emission. We quantified the spectral diffusion by analyzing the time-resolved PL intensity under resonant excitation. The blue dashed line shows the threshold of 400 cps used to define on and off times under resonant excitation.

Over 10 min, the emitter was excited coherently for ∼80 s (τ_{on}), and the rest of the time the emitter was off resonance due to spectral diffusion. The τ_{on}/τ_{off} ratio of 0.13 shows that the emitter is on resonance 13% of the time. Note that we did not observe a power dependence of the spectral diffusion, and blinking dynamics was recorded using a much lower excitation power of ∼500 nW. The spectral diffusion may occur due to charge fluctuations induced by the laser field, and can be mitigated dynamically using Stark shift tuning [16]. However, even under these conditions, given the high brightness and the high Debye–Waller factor of these emitters, one would expect more than 1×10^5 photons/s on resonance from the ZPL, which is suitable for practical devices.

In conclusion, we demonstrated that a novel quantum system—single emitters in GaN—can be excited coherently. Near FT-limited linewidths of ∼250 MHz were observed and spectral diffusion was quantified. Our results pave the way for integrated quantum photonic circuitry based on GaN and provide important information on emerging quantum emitters in this material.

Funding. Australian Research Council (ARC) (DP180100077, DE180100810); Office of Naval Research Global (ONRG) (N62909-18-1-2025); Engineering and Physical Sciences Research Council (EPSRC) (EP/M011682/1).

REFERENCES

1. X.-L. Chu, S. Götzinger, and V. Sandoghdar, Nat. Photonics **11**, 58 (2017).
2. T. D. Ladd, F. Jelezko, R. Laflamme, Y. Nakamura, C. Monroe, and J. L. O'Brien, Nature **464**, 45 (2010).
3. P. Senellart, G. Solomon, and A. White, Nat. Nanotechnol. **12**, 1026 (2017).
4. A. Sipahigil, R. E. Evans, D. D. Sukachev, M. J. Burek, J. Borregaard, M. K. Bhaskar, C. T. Nguyen, J. L. Pacheco, H. A. Atikian, C. Meuwly, R. M. Camacho, F. Jelezko, E. Bielejec, H. Park, M. Lončar, and M. D. Lukin, Science **354**, 847 (2016).
5. N. Kalb, A. A. Reiserer, P. C. Humphreys, J. J. W. Bakermans, S. J. Kamerling, N. H. Nickerson, S. C. Benjamin, D. J. Twitchen, M. Markham, and R. Hanson, Science **356**, 928 (2017).
6. S. Castelletto, B. C. Johnson, V. Ivády, N. Stavrias, T. Umeda, A. Gali, and T. Ohshima, Nat. Mater. **13**, 151 (2014).
7. D. J. Christle, A. L. Falk, P. Andrich, P. V. Klimov, J. U. Hassan, N. T. Son, E. Janzen, T. Ohshima, and D. D. Awschalom, Nat. Mater. **14**, 160 (2015).
8. M. Kianinia, B. Regan, S. A. Tawfik, T. T. Tran, M. J. Ford, I. Aharonovich, and M. Toth, ACS Photon. **4**, 768 (2017).
9. T. T. Tran, M. Kianinia, M. Nguyen, S. Kim, Z.-Q. Xu, A. Kubanek, M. Toth, and I. Aharonovich, ACS Photon. **5**, 295 (2017).
10. X. He, H. Htoon, S. K. Doorn, W. H. P. Pernice, F. Pyatkov, R. Krupke, A. Jeantet, Y. Chassagneux, and C. Voisin, Nat. Mater. **17**, 663 (2018).
11. M. J. Holmes, K. Choi, S. Kako, M. Arita, and Y. Arakawa, Nano Lett. **14**, 982 (2014).
12. A. M. Berhane, K. Y. Jeong, Z. Bodrog, S. Fiedler, T. Schroder, N. V. Trivino, T. Palacios, A. Gali, M. Toth, D. Englund, and I. Aharonovich, Adv. Mater. **29**, 1605092 (2017).
13. Y. Zhou, Z. Wang, A. Rasmita, S. Kim, A. Berhane, Z. Bodrog, G. Adamo, A. Gali, I. Aharonovich, and W.-B. Gao, Sci. Adv. **4**, eaar3580 (2018).
14. R. Datta, M. J. Kappers, M. E. Vickers, J. S. Barnard, and C. J. Humphreys, Superlattices Microstruct. **36**, 393 (2004).
15. S. Buckley, K. Rivoire, and J. Vuckovic, Rep. Prog. Phys. **75**, 126503 (2012).
16. V. M. Acosta, C. Santori, A. Faraon, Z. Huang, K. M. C. Fu, A. Stacey, D. A. Simpson, K. Ganesan, S. Tomljenovic-Hanic, A. D. Greentree, S. Prawer, and R. G. Beausoleil, Phys. Rev. Lett. **108**, 206401 (2012).