Robust Multicolor Single Photon Emission from Point Defects in Hexagonal Boron Nitride

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Abstract: We demonstrates engineering of quantum emitters in hBN multi-layers using either electron beam irradiation or annealing. The defects exhibit a broad range of multicolor room-temperature single photon emissions across the visible and the near-infrared ranges.

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1. Full Abstract

Artificial atomic systems in solids are widely considered the leading physical system for a variety of quantum technologies, including quantum communications, computing and metrology.[1, 2] Room temperature quantum emitters have, however, thus far been observed only in wide bandgap semiconductors such as diamond[3] and silicon carbide,[4] nanocrystal quantum dots,[5-7] and most recently carbon nanotubes[8]. In previous reports we have demonstrated for the first time quantum emission from hexagonal boron nitride (hBN) monolayer,[9] multilayer,[9] and bulk.[10] The single photon nature originates from localized optically active defect centers embedded within hBN lattice. To further understand more about these color centers, we focus on fabrication techniques and optical properties of these emitters.

In this work, we show that single emitters in hBN can be engineered using either annealing or electron beam irradiation. While the use of heat gives rise to high yield of generation of optically active centers, the spatial distribution of the emitters are relatively random. On the other hand, by using electron beam irradiation with existence of low-concentration of water vapor, we managed to generate defect centers in a more predetermined fashion. We characterize the emitters and report an interesting phenomenon, namely, narrowband multicolor single photon emission from a 2D material. While known color centers and standard quantum dots (of a given, fixed chemical composition) luminesce at a particular wavelength or over a narrow spectral range, we show that defects in hBN multilayers can emit over a broad range spanning over 200 nm. We also propose that there are two groups of emitters based on their unique optical and photophysical properties. With density functional theory (DFT) calculation, it is proposed that the shifts in emission energy of zero-photon lines (ZPLs) in individual group of emitters could be due to strain in hBN lattice. We also show that the emitters withstand various aggressive annealing treatments in reactive gaseous environments, namely, H_2, O_2 and NH_3, which do not change their spectral properties. Our results pave the way to robust, room-temperature quantum photonic devices that employ color centers in hBN as key building blocks.[11]

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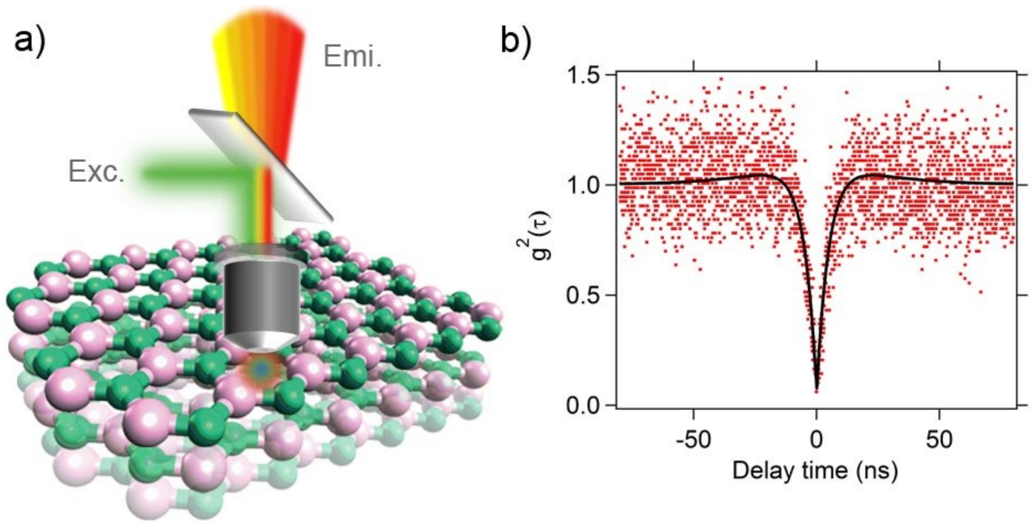

Fig. 1 (a) Schematic showing narrow-band multicolor single photon emission from multilayer hBN. (b) A representative second order autocorrelation function providing an explicit proof for quantum emission from multilayer hBN.

2. Summary

In conclusion, we present two robust methodologies to engineer room-temperature multicolor single photon emission, based on annealing and electron beam irradiation. Moreover, we show that the emitters are stable even after annealing in harsh gaseous environments such as oxygen, hydrogen, and ammonia. By analyzing spectral features of these emitters, we could infer that there are at least two groups of defects. Although the emitters in the two groups exhibit significant differences in their spectral characteristics, they share similar local phonon energies and therefore are likely to have similar chemical structure. Our work may help open up possibilities for employing quantum emitters in 2D materials for emerging applications in nanophotonics and nanoscale sensing devices.

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4. References

1. O'Brien, J. L.; Furusawa, A.; Vučković, J. Photonic Quantum Technologies. *Nature Photon.* 2009, 3, 687-695.
2. Awschalom, D. D.; Bassett, L. C.; Dzurak, A. S.; Hu, E. L.; Petta, J. R. Quantum Spintronics: Engineering and Manipulating Atom-Like Spins in Semiconductors. *Science* 2013, 339, 1174-1179.
3. Aharonovich, I.; Neu, E. Diamond Nanophotonics. *Adv. Opt. Mater.* 2014, 2, 911-928.
4. Castelletto, S.; Johnson, B. C.; Ivády, V.; Stavrias, N.; Umeda, T.; Gali, A.; Ohshima, T. A Silicon Carbide Room-Temperature Single-Photon Source. *Nature Mater.* 2014, 13, 151-156.
5. Mangum, B. D.; Wang, F.; Dennis, A. M.; Gao, Y.; Ma, X.; Hollingsworth, J. A.; Htoon, H. Competition between Auger Recombination and Hot-Carrier Trapping in Pl Intensity Fluctuations of Type Ii Nanocrystals. *Small* 2014, 10, 2892-2901.
6. Pisanello, F.; Lemenager, G.; Martiradonna, L.; Carbone, L.; Vezzoli, S.; Desfonds, P.; Cozzoli, P. D.; Hermier, J. P.; Giacobino, E.; Cingolani, R.; De Vittorio, M.; Bramati, A. Non-Blinking Single-Photon Generation with Anisotropic Colloidal Nanocrystals: Towards Room-Temperature, Efficient, Colloidal Quantum Sources. *Adv. Mater.* 2013, 25, 1974-1980.
7. Michler, P.; Imamoglu, A.; Mason, M. D.; Carson, P. J.; Strouse, G. F.; Buratto, S. K. Quantum Correlation among Photons from a Single Quantum Dot at Room Temperature. *Nature* 2000, 406, 968-970.
8. Ma, X.; Hartmann, N. F.; Baldwin, J. K.; Doorn, S. K.; Htoon, H. Room-Temperature Single-Photon Generation from Solitary Dopants of Carbon Nanotubes. *Nat. Nanotechnol.* 2015.
9. Tran, T. T.; Bray, K.; Ford, M. J.; Toth, M.; Aharonovich, I. Quantum Emission from Hexagonal Boron Nitride Monolayers. *Nat. Nanotechnol.* 2016, 11, 37-41.
10. Tran, T. T.; Zachreson, C.; Berhane, A. M.; Bray, K.; Sandstrom, R. G.; Li, L. H.; Taniguchi, T.; Watanabe, K.; Aharonovich, I.; Toth, M. Quantum Emission from Defects in Single-Crystalline Hexagonal Boron Nitride. *Phys. Rev. Appl.* 2016, 5, 034005.
11. Tran, T. T.; Elbadawi, C.; Totonjian, D.; Lobo, C. J.; Grosso, G.; Moon, H.; Englund, D. R.; Ford, M. J.; Aharonovich, I.; Toth, M. Robust Multicolor Single Photon Emission from Point Defects in Hexagonal Boron Nitride. *ACS Nano* 2016, 10, 7331-7338.