Photophysical Characterization of Quantum Emitters in Hexagonal Boron Nitride (h-BN)

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Abstract: Optical emission and Raman characteristics of defect centers in h-BN have been systematically investigated within different dielectric environments. These studies provide new insights into better creation and engineering of h-BN based quantum emitters.
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1. Introduction

Single-photon emitters (SPEs) are the central building blocks for quantum circuits, which are envisaged to revolutionize information processing, communication, and sensing technologies. Thanks to the advances in material science and nanotechnology, a proliferation of defect-related SPEs has been achieved in wide-bandgap (WBG) materials [1]. Among these materials, hexagonal boron nitride (h-BN) has rapidly emerged as an attractive photonic platform and sparked growing research to unveil the quantum behavior associated with its two-dimensional (2D) structure [2]. The ultrawide bandgap of h-BN (5.9 eV) has been demonstrated capable of hosting optically active defect centers that strongly emit single photons even at room temperature. The planar geometry and in-plane confinement of optical dipoles offer great potential in integration with other photonic structures. SPEs in these 2D layers also possess remarkably tunability, and exquisitely high responsivities to the events on surface, compared with their counterparts buried inside conventional 3D crystals (*e.g.*, diamond, silicon carbide). Albeit, susceptibility to disturbances and spectral diffusion are detrimental to the purity, indistinguishability, and stability of SPEs. Therefore, it is highly desirable to explore, understand, and attain deterministic activation and control of SPEs in h-BN crystals.

2. Experimental Results and Discussion

The h-BN samples are prepared via a suite of completely dry exfoliation and transfer methods, as illustrated in Fig. 1a. Multilayer h-BN flakes are mechanically exfoliated from a high-quality bulk h-BN crystal and pressed onto polydimethylsiloxane (PDMS) stamps. After exfoliation, the h-BN nanosheets are transferred onto various patterned substrates, including bare silicon (Si) and silicon dioxide on silicon (SiO_2-on-Si). As-prepared samples consist of supported and suspended h-BN regions (Fig. 1b-1d), serving as excellent platforms to explore the influence of dielectric environment and local strain on the quantum emission properties. Prior to the optical spectroscopic characterization, high-temperature annealing at 850 °C under 1 Torr of nitrogen is performed to activate the defect centers and to desorb any possible surface contaminants.

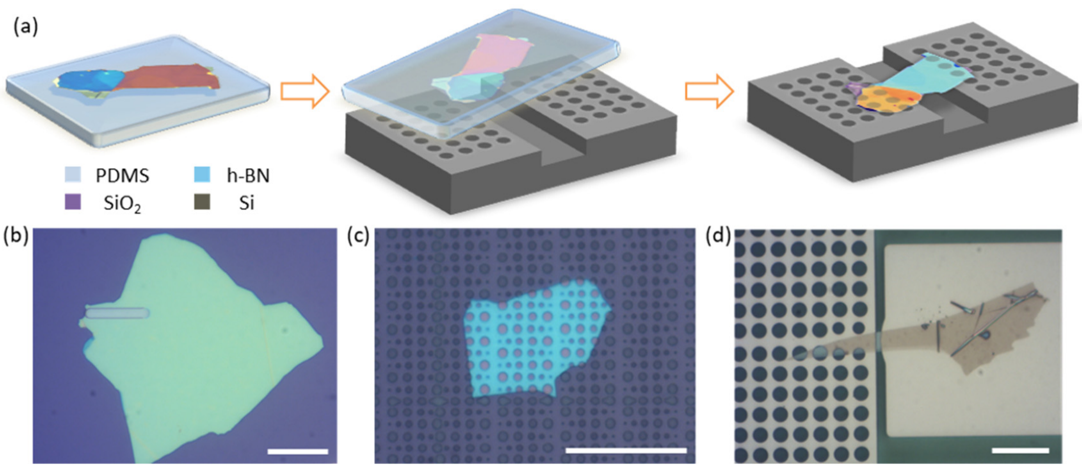

Fig. 1. (a) Real color and inverted color images of an h-BN flake, and schematic illustration of the PDMS-assisted dry transferring of h-BN crystals onto the patterned substrate. Optical microscopy images of h-BN samples on (b)-(c) SiO_2-on-Si and (d) Si substrate. Scale bar: 20 µm.

To better understand the crystal quality and defect behaviors in these h-BN samples, we use spectroscopic tools to probe the vibrational modes and photoluminescence (PL) in the materials. Our system consists of a green laser (532 nm), high-magnification optical objectives (50× or 100×), and a spectrometer (Horiba iHR550 or Princeton Instruments 2500) with a detector sensitive to record the weak scattered light signals for Raman and single photon measurement (Fig. 2a). The customized microscopy system is capable of acquiring detailed and local information with a high spatial resolution (~1 µm) and high collection efficiency. Combined with a motorized stage and digital imaging, it can reveal both the fluorescence image and PL/Raman mapping over a certain spatial range. Correlation measurements are performed by using the Hanbury Brown and Twiss interferometry setup at zero delay time to further verify the single photon nature of the emission process. Without specification, all the measurements are conducted at room temperature.

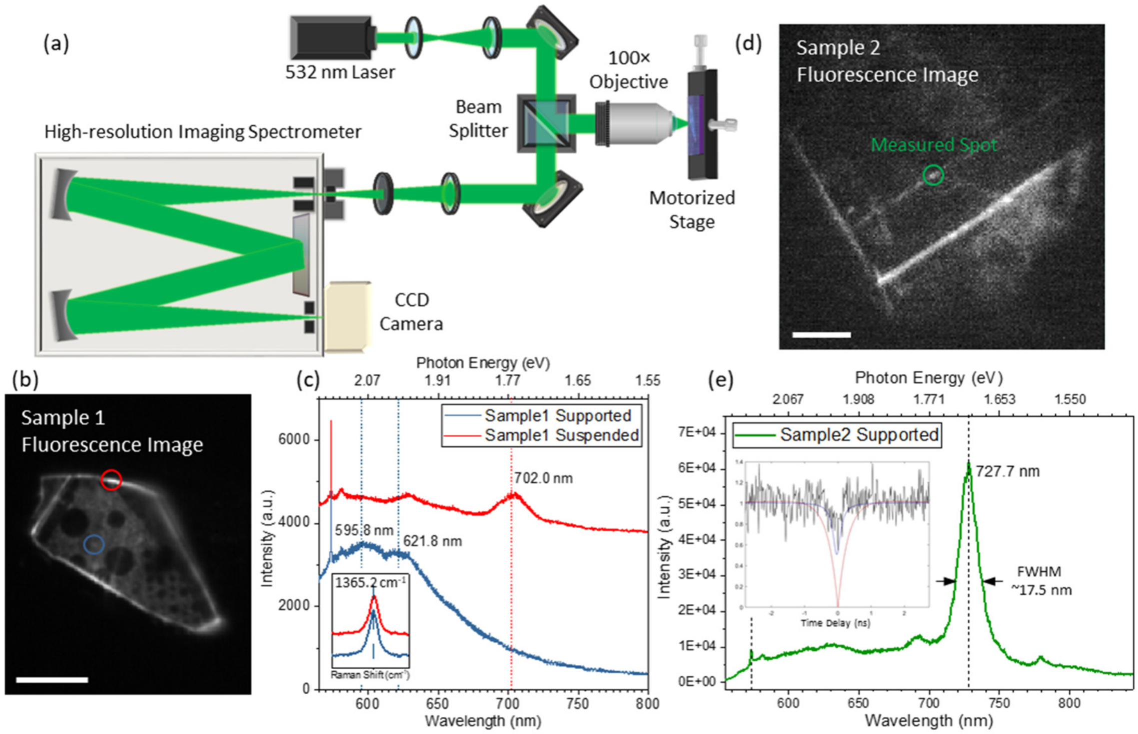

Fig. 2. (a) Schematic illustration of the optical spectroscopy system. (b)-(c) Fluorescence image and emission spectra of h-BN on perforated SiO_2-on-Si substrate. (d)-(e) Fluorescence image, spectrum, and correlation curves of h-BN on unpatterned SiO_2-on-Si substrate. Scale bar: 10 µm.

Panels (b)-(e) of Fig. 2 exemplify the optical signatures of defect centers in h-BN. For the sample shown in Fig. 2b (Sample 1), the supported and suspended areas exhibit different emission spectral profiles, with emission center around 600 nm and 700 nm, respectively (Fig. 2c). With the 532nm laser excitation, the sharp peak emerging at ~573 nm (~1365 cm^{-1}) can be assigned as transverse optical (TO) mode of h-BN Raman peak. A slight red-shift can be resolved in the suspended region compared to the TO mode position of the supported area. A more pronounced emission peak around 727 nm can be observed in Sample 2 (Fig. 2d), with a narrow linewidth of 17.5 nm. The correlation measurement result from Sample 2 is shown as the black curve in the inset of Fig. 2e. The fitted curve in blue is obtained using a Lorentz function, and the ideal correlation curve in red is fitted using a three-level model. With a true single-photon source, the $g^2(\tau)$ curve dips below 0.5 at zero delay time, which means the possibility of having single photon is larger than the possibility of having two at the same time.

3. Conclusion

We chave haracterized the defect centers in h-BN by using Raman and photoluminescence (PL) spectroscopy. A better understanding of the material and photophysical properties has been developed, which will serve as a stepping stone toward engineering h-BN quantum devices for future computation and communication applications.

4. References

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