

BORON NITRIDE WORKSHOP 2024

May 21st - 23rd

UTS Aerial
Function Centre



University of
Technology Sydney,
Australia

WELCOME TO THE BORON NITRIDE WORKSHOP 2024



Dear Friends and Colleagues,

Welcome to Sydney, and to the 2nd Boron Nitride Workshop. We are excited to see our community grow, and thank you for taking part in this amazing journey.

Following the successful launch of the BNW series in Montpellier in 2023, this year, the scope of the workshop has expanded to include a dedicated session on cubic BN and energy applications. We are of course maintaining the broader scope of the workshop to include all key areas of research surrounding BN – spanning electronics, polaritonics, lighting, imaging, quantum, biology, materials science and theory.

We are also delighted to welcome attendees from 16 countries and we hope to see our global community grow in years to come. We hope you enjoy your time in Sydney and engage with the BN scientific community.

-Igor, Josh and Guillaume

UTS acknowledges the Gadigal people of the Eora Nation, the Boorooberongal people of the Dharug Nation, the Bidiagal people and the Gamaygal people, upon whose ancestral lands our university stands. We would also like to pay respect to the Elders both past and present, acknowledging them as the traditional custodians of knowledge for these lands.

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INVITED SPEAKERS

FRANCISCO MUNOZ

Universidad de Chile, Santiago, Chile

Carbon-based single photon emitters in hBN and van der Waals heterostructures

JIEUN LEE

Seoul National University, Korea

Electrical control of light emission from h-BN single photon sources

YING IAN CHEN

Deakin University, Waurn Ponds, Australia

Boron Nitride Nanotubes and Nanosheets: from Scientific Discoveries to Commercialisation

SIYUAN DAI

Auburn University (USA)

Engineering phonon polaritons in hBN van der Waals structures

SIDDHA PIMPUTKAR

Lehigh University, USA

Growth of Bulk Boron Nitride

YOUNG DUCK KIM

Kyung Hee University, Seoul, South Korea

Manipulation of carbon color centers in hexagonal boron nitride for efficient deep ultraviolet light emission

LUDGER WIRTZ

University of Luxembourg

Signatures of exciton-phonon coupling in spectroscopy of layered boron nitride

KAZUYUKI HIRAMA

NTT Basic Research Laboratories

c-BN epitaxial growth mechanism in ion-beam-assisted MBE

INVITED SPEAKERS

TOMOKI MACHIDA

Institute of Industrial Science, University of Tokyo

Hexagonal boron nitride in van der Waals junctions of two-dimensional materials: substrate, capping layer, tunnel barrier, and potential modulator

PENGFEI YANG

HK University

Controllable growth of uniform multilayer hexagonal boron nitride on metals and insulators

VIKTOR IVÁDY

Eötvös Loránd University

Computational exploration of hBN defects: insights into topological defects, spin qubits, and the origins of visible color centers

WEN-HAO CHANG

National Yang Ming Chiao University

CVD-grown hBN for 2D transistors and quantum emitters

KAIHUI LIU

Peking University

Optical Crystals of Two-dimensional Rhombohedral Boron Nitride

ANDRZEJ WYSMOLEK

University of Warsaw

MOVPE growth and applications of layered boron nitride

STEFAN MAIER

Monash University

Tunability and applications of hBN metasurfaces

VALENTYN S. VOLKOV

Emerging Technologies Research Center

Hexagonal Boron Nitride Nanophotonics: Ultraviolet Transparency, High Refractive Index and Optical Anisotropy

ORAL PRESENTATIONS

SIMONAS KROTKUS

Interfacial engineering for wafer scale synthesis of multilayer sp^2 -BN films

SEOKHO MOON

Wafer-Scale AA-Stacked Hexagonal Boron Nitride Grown on GaN Substrate

RUOTIAN GONG

Enhancing Coherence Properties of Spin Defects in hBN

SHENG-SHONG WONG

The electronic band structure of mono-, bi-, and trilayer h-BN

TONGCANG LI

Quantum sensing with single spin defects in boron nitride nanotubes

KENTO SASAKI

Nitrogen isotopes effects on hBN quantum sensor

JONATHAN BRADFORD

Molecular beam epitaxy of carbon-doped hexagonal boron nitride on HOPG: Insights into the atomic structure of single photon emitters

GYUNA PARK

Room temperature electroluminescence from isolated colour centres in van der Waals semiconductors

CAMILLE MAESTRE

Searching for diffuse defects in millimeter-sized h-BN crystals

ORAL PRESENTATIONS

GUILLAUME CASSABOIS

Optical and spin properties of boron-vacancy centers in few-layer thick hexagonal boron nitride

KRISTINA MALINOWSKI

Photon statistics analysis of h-BN quantum emitters with pulsed and continuous-wave excitation through Mandel Q

LESLEY SPENCER

Monolithic Integration of Single Quantum Emitters in hBN Bullseye Cavities

JOSHUA CALDWELL

Ultrafast Thermal Dissipation via Surface Phonon Polaritons

ISLAY ROBERTSON

Spin properties of visible emitters in hBN and their applications

EVELINE MAYNER

Optical Readout of Redox Reaction via hBN Surface Emitters

GEORGE BEPETE

Chemical intercalation, exfoliation, and functionalization of hBN materials

MYUNGSOO KIM

Hexagonal Boron Nitride Memristors and RF Switches

POSTER PRESENTATIONS

SERGEI NEDIC

Electron Beam Restructuring of Quantum Emitters in Hexagonal Boron Nitride

ANAND KUMAR

Solid states quantum emitters in wide band gap materials for quantum technology applications

QIRAN CAI

Thermal property and applications of boron nitride nanosheets

GUILLAUME CASSABOIS

What is the nature of the UV color center emitting at 300 nm in hexagonal boron nitride ?

CHRISTOPHER J. MELLOR

Single photon emitters created by intentional carbon doping of hexagonal boron nitride grown on sapphire by high-temperature molecular beam epitaxy

KABILAN SRIPATHY

QUICK3 – Towards satellite-based quantum communication, and fundamental physics tests in microgravity

ROTEM MALKINSON

Systematically creating boron vacancies in bulk exfoliated hexagonal boron nitride flakes using focused ion beam

PAUL KONRAD

Spin-1 Quantum Sensors in hBN: Intersystem Crossing Relaxation of the Metastable State and Irradiation Protocol

JAKUB ROGOŹA

Conductivity induced by post growth annealing of boron nitride grown by MOVPE

SONACHAND ADHIKARI

hBN-Enabled Flexible GaN Photodetector

POSTER PRESENTATIONS

PRAGYA JOSHI

Carbon Migration and Single Photon Emission in Electron Irradiated Hexagonal Boron Nitride Flakes

JAKUB IWAŃSKI

Polytype Identification in MOVPE Grown sp²-BN Using Ultraviolet Defect Photoluminescence

QIRAN CAI

Boron nitride nanosheet aggregates for enhanced acoustic energy harvesting

ROY STYLES

The effect of electric fields on visible spin defects in hBN

DOMINIC SCOGNAMIGLIO

Controlling and stabilizing the Charge State of Spin Defects in hBN

JONG SUNG MOON

Fiber-integrated quantum sensors using color centers with optimal cavity interface

BINDU BINDU

Quantum Sensing and Imaging of van der Waals Ferromagnet using Nitrogen-Vacancy Centers

GALYA HAIM

Exploring methods for creation of Boron-vacancies in hexagonal Boron Nitride exfoliated from bulk crystal

MOMOKO ONODERA

Evaluation of hexagonal boron nitride in van der Waals junctions of 2D materials

JULIETTE PLO

Nitrogen isotope effects on hexagonal boron nitride

POSTER PRESENTATIONS

MIN-JAE MAENG

Transport band gap measurement of large-area hBN by using direct and inverse photoemission spectroscopy

RICHARD ESCALANTE

Sensitivity Optimization of Boron Vacancy Centers in Hexagonal Boron Nitride

ANDREW BELING

Toward Nanoscale NMR Spectroscopy using the Boron Vacancy Quantum Defect in hBN

GUILLAUME CASSABOIS

Spatially-resolved UV-C emission in epitaxial monolayer boron nitride

MINHYUN CHO

Remote moire effect engineering using the twisted hBN

YOUNGJAE KIM

High electric field vertical tunneling transports in hexagonal boron nitride

HEEYEON LEE

Hexagonal boron nitride surface engineering for remote modulation doping

SHIH-CHU LIN

Defect engineering in CVD-Grown Hexagonal Boron Nitride for Quantum Photonic Applications

SEUNGMIN PARK

High-efficiency deep ultraviolet emitting from hexagonal boron nitride heterostructure

NILS BERNHARDT

UV defect emitters in thin hBN

TAKASHI TANIGUCHI

TBD

POSTER PRESENTATIONS

HELEN ZENG

Hexagonal Boron Nitride-Based Quantum Key Distribution with Room Temperature Single Photon Emission

BENJAMIN VINCENT WHITEFIELD

Magnetic Field Sensitivity Optimization of Negatively Charged Boron Vacancy Defects in hBN

KARIN YAMAMURA

Plasmonic lattices-assisted emission enhancement and optimized creation of blue colour centre in hBN

JAKE HORDER

Resonant Spectroscopy of B-Center Quantum Emitters in hBN

MADELINE HENNESSEY

Towards Boron Nitride Nanotube Optical Emitters in Sensing Applications

IVAN ZHIGULIN

Photodynamics of electrically driven isolated colour centres in van der Waals semiconductors

RYAN KOWALSKI

Correlating Defect Emission with Infrared Near-Field Imaging in Strained Hexagonal Boron Nitride

WEI LIU

Coherent control of an ultrabright single spin in hexagonal boron nitride at room temperature

PIRAN RAVICHANDRAN KIDAMBI

Nanoporous atomically thin ceramic membranes for energy and healthcare

HAYOUNG KO

Growth of wafer-scale, high-quality, and uniform multilayer hBN film on liquid Fe₂B for high-performance of 2D heterostructure

Carbon-based single photon emitters in hBN and van der Waals heterostructures

Francisco Munoz

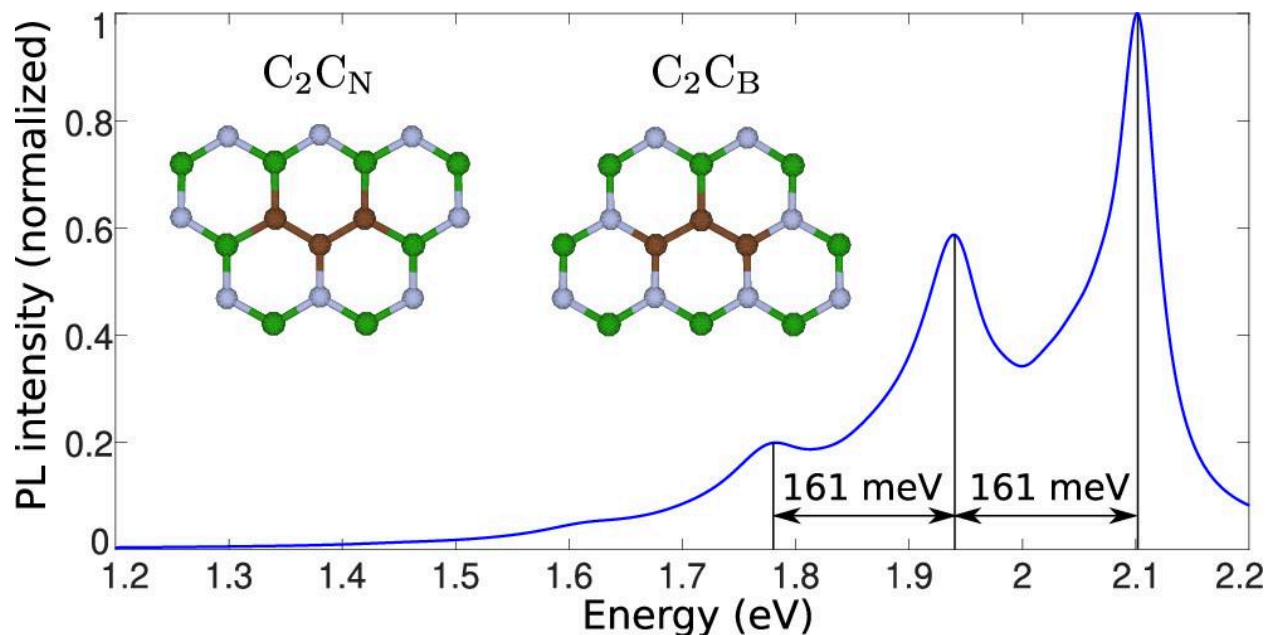
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Hexagonal boron nitride (hBN) has been discovered to host natural single photon emitters, which can also be induced through irradiation, emitting across the entire visible spectrum. Despite variations in emission energy, a characteristic phonon-sideband is found, featuring phonon replicas at approximately 160-170 meV.

This presentation delves into the identification of C substitutional defects as the source of single photon emission, employing density functional theory calculations. These defects may manifest as individual clusters of C atoms or more intricate structures. Their electronic structure resembles a donor-acceptor system, promoting a singlet or doublet ground state, although the existence of triplet or higher spin C-based defects is also observed.

Furthermore, time permitting, the discussion will extend to the manipulation of emission properties through the integration of van der Waals heterostructures.



Electrical control of light emission from h-BN single photon sources

Jieun Lee (Seoul National University)

Abstract:

Single photon sources are fundamental resources of quantum optics and quantum information technologies. The emergence of quantum emission in hexagonal boron nitride (h-BN) has triggered tremendous interests in atomically thin 2D material based single photon sources. For the full exploitation of 2D single photon sources for quantum technologies, however, the ability to control each atomic defect individually is crucial. In this talk, we introduce methods to generate and manipulate quantum emission from 2D h-BN crystals by using an external voltage. In the first part, we describe the electric field induced energy tuning of quantum emitters in h-BN. By applying an external field to graphene/h-BN/graphene stacked heterostructures, we observed a diverse trail of Stark shifts, from which various information about the crystallographic nature of atomic defects is extracted. In the second part, we discuss the charge state manipulation of quantum emitters in h-BN by using an external voltage. By inducing the electron transfer from adjacent charge reservoirs, we observed the luminescence brightening of quantum emitters in h-BN and the precision measurement of charge transition level allows the identification of the defect structure. These results will be beneficial for the development of on-chip photonic quantum information devices using 2D materials.

Boron Nitride Nanotubes and Nanosheets: from Scientific Discoveries to Commercialisation

Alfred Deakin Professor Ying Ian Chen

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The field of boron nitride (BN) nanomaterials encompasses nanotubes and nanosheets. While BN nanotubes share a similar structure with carbon nanotubes, they possess distinct chemical and thermal properties that enable special applications in composite materials, aerospace, and new batteries. My team started the research in BN nanotubes because of the discovery of a novel production method of BN nanotubes utilizing high-energy ball milling and thermal annealing processes, leveraging mechanochemistry to facilitate the growth of two-dimensional (2D) structures [1]. This production method has been successfully commercialized by BNNT Technology Ltd, a spin-off company, and BN nanotubes are currently employed in lithium-sulfur batteries by LiS Energy Ltd, an Australian company listed on the stock market.

On the other hand, BN nanosheets possess a hexagonal structure akin to graphene but exhibit unique properties and applications distinct from graphene. Our current research focuses on mass synthesis techniques, novel properties, and applications in gas storage, batteries and other fields. This presentation will showcase our journey from fundamental discoveries to commercialization endeavors, highlighting recent research findings on the exceptional mechanical properties [2], effective oil-water separation capabilities [3], the BN nanosheet membranes in fluidic devices [4,5], their outstanding thermal conductivity [6], their ability to enhance the cycling life of Li-S batteries [7,8], the development of new catalysts [9], as well as their potential for gas storage and separation [10]. For more information, please visit <http://www.deakin.edu.au/about-deakin/people/ying-ian-chen>.

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Engineering phonon polaritons in hBN van der Waals structures

Siyuan Dai

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The manipulation of light at small scales is one of the ultimate goals of modern optics. For this purpose, polaritons—hybrid light-matter waves propagating in a confined length scale—are typically involved. Recent results of polaritons in van der Waals materials reveal a series of advances attributed to the unique physical properties in reduced dimensions and the configurability through van der Waals engineering. In this talk, I will describe the unique characteristics of photolattice waves—phonon polaritons—in hBN by van der Waals engineering. By stacking, patterning, and isotope heterostructuring, phonon polaritons in hBN van der Waals structures exhibit dynamic and reversible tunability, exotic nano-light wavefronts, mode conversions, and new energy-momentum dispersions.

Growth of Bulk Boron Nitride

Siddha Pimputkar

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Boron nitride (BN) is a fascinating material offering extreme and unique material properties that can be leveraged in a range of potential applications including (opto)electronic and quantum devices. Availability of high-quality, large-area and volume, single-crystal material would provide a significant boon to the community facilitating or enabling development of devices. BN has been successfully grown using high pressure, high temperature presses and precipitation from a solvent on the order of a few millimeters. While these methods have yielded high-quality material, scaling of these approaches is challenging motivating the search for additional bulk synthesis methods. This talk will present foundational work being performed in the pursuit of bulk BN growth using two industrial scalable methods: the ammonothermal method and a flux-based approach.

The ammonothermal method utilizes supercritical ammonia at temperatures and pressures around 400—600 °C and 100—250 MPa, respectively. One or more mineralizers are added to the solution to enhance the solubility of BN. This talk will demonstrate temperature-dependent solubility of BN in basic ammonothermal solutions containing alkali [1] and/or alkaline earth metals. Initial crystal growth experiments were performed using sodium as the mineralizer yielding the spontaneous nucleation and growth of sub-mm sized hexagonal and rhombohedral BN.

The flux-based method uses a solvent exhibiting exceptionally high solubility of nitrogen and boron. A dedicated growth system has been developed and initial growth runs have demonstrated the solubility and growth of BN from solution. Preliminary growth campaigns have successfully demonstrated the growth of $\sim >0.1$ mm sized BN crystals after 1-2 days.

This work has been supported by NSF DMR 1832824, NSF CAREER 2046468, and ARL DEVCOM UWBG RF Electronics Center.

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Manipulation of carbon color centers in hexagonal boron nitride for efficient deep ultraviolet light emission

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Color centers in wide-bandgap semiconductors such as hexagonal boron nitride (hBN) play an important role in quantum technologies such as single photon emitters and spin qubits. Especially carbon color centers in hBN, substituted carbon atoms at boron or nitride lattice, emerged as the atomic origin of several single photon emitters in several emission energies due to the robust deep-level trap states inside the bandgap. Here, I will present the creation and manipulation of carbon color centers in hBN and their deep ultraviolet (UV) emission properties, such as photoluminescence (PL) and electroluminescence (EL). Carbon color centers in hBN with van der Waals heterostructures exhibit robust tunneling behaviors with the highest breakdown electric fields, which is attributed to carbon atoms in hBN being the most stable defect configuration with the largest binding energy. Furthermore, I will also present the efficient electrically driven deep UV light emission from carbon-doped hBN van der Waals heterostructures with efficient charge carrier injections via trap states. These results pave the way for the promising developments of a highly efficient solid-state DUV light source and solid-state quantum emitter at the nanoscale.

Signatures of exciton-phonon coupling in spectroscopy of layered boron nitride

Ludger Wirtz

Department of Physics and Materials Science

University of Luxembourg

I will present a review on the latest developments in theoretical spectroscopy of single-layer, few-layer and bulk boron nitride. Albeit its structural simplicity, the luminescence spectra of BN feature a surprising complexity.

Due to its layered structure and large band gap, excitonic effects are strongly pronounced in this material. A proper calculation of excitonic states - both with zero momentum and with finite momentum - is thus the starting point for exploring the various spectra (absorption, luminescence, Raman) of BN. We have developed a theoretical and computational framework to take into account exciton-phonon coupling, i.e., the coupling of electronic excitations to lattice vibrations. A group-theoretical analysis greatly simplifies the understanding of which couplings are possible and which are not.

I will present how phonon-assisted luminescence spectroscopy allows to distinguish different stackings (hexagonal, rhombohedral, Bernal) of BN. Furthermore, I will present intriguing inter-layer exciton-phonon coupling effects at interfaces of BN with transition-metal dichalcogenides, rendering "dark" phonon modes Raman active. Last but not least, I will talk about the defect signatures in the optical spectra of BN.

It is a pleasure to acknowledge the contributions of various collaborators to this work: Sven Reichardt and Muralidhar Nalabothula (Resonant Raman Spectroscopy), Fulvio Paleari, Matteo Zanfronini, Daniele Varsano (phonon-assisted luminescence), Davide Sangalli (Symmetries and exciton-phonon coupling) and Henry Fried (defect signatures).

Controllable growth of uniform multilayer hexagonal boron nitride on metals and insulators

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ABSTRACT: Multilayer hexagonal boron nitride (h-BN), a 2D crystalline insulator with a wide band gap (~ 6 eV), has shown great potential in versatile applications for 2D electronic and optoelectronic devices due to its atomic flatness and the absence of dangling bonds on its surface. However, the controllable synthesis of uniform and scalable multilayer h-BN on metals or insulating substrates via the chemical vapor deposition is still challenging, due to the lack of understanding on the multilayer growth mechanisms. In this study, we introduce the controllable growth strategies for the uniform multilayer h-BN films by using single-crystal Ni(111) and sapphire as templates. Experimental results and theoretical calculations reveal that, the uniform multilayer h-BN grown on Ni substrate is via a vapor-liquid-solid (VLS) process, where a heterogeneous Ni-B liquid layer on the Ni(111) substrate serves as a reservoir to supply B and N sources for multilayer h-BN growth. And in a sharp contrast, the growth h-BN on sapphire substrate is a vapor-solid (VS) process, where the catalytic inert surface of sapphire substrate plays a minor role in the feeding process of BN species, and is mainly functioned as a support for h-BN growth. Furthermore, the potential applications of these uniform multilayer h-BN films in large-scale high-density memristive arrays are also demonstrated.

Hexagonal boron nitride in van der Waals junctions of two-dimensional materials: substrate, capping layer, tunnel barrier, and potential modulator

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Recent advances in transfer techniques of atomic layers have enabled one to fabricate van der Waals junctions of two-dimensional (2D) materials. Hexagonal boron nitride (h-BN) is the insulating 2D material that has been widely used as an atomically flat substrate, a gate insulator, a tunnel barrier, and a moiré potential modulator. Here, we present our recent experiments on fabrication and quantum transport of van der Waals junctions of h-BN and various 2D materials.

First, we utilize h-BN as a tunnel barrier for resonant tunneling between quantized subbands. We demonstrate van der Waals double quantum well (vDQW) devices based on few-layer WSe₂ quantum wells and a few-layer h-BN tunnel barrier. Due to the strong out-of-plane confinement, WSe₂ exhibits quantized subband states at the Γ point in its valence band. Here, we report resonant tunneling and negative differential resistance in vDQW at room temperature owing to momentum- and energy-conserved tunneling between the quantized subbands.

Next, we use h-BN as a potential modulator to study van Hove singularity (vHS) in graphene/h-BN moiré superlattice. The thermoelectric voltage for the vHS at the low energy side of the hole-side secondary Dirac point exhibited significant magnetic field-induced valley splitting with an effective g-factor of approximately 130. This is attributed to the emergence of an orbital magnetic moment at the second vHS.

Furthermore, we discuss the defect-assisted resonant tunneling spectroscopy through carbon-doped h-BN, the influence of carbon-rich domain in h-BN, the quality of h-BN crystals synthesized at atmospheric pressure as insulating layers in vdW heterostructures, and 3D manipulation technique of 2D material flakes.

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Computational exploration of hBN defects: insights into topological defects, spin qubits, and the origins of visible color centers

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In this presentation, I delve into the diverse aspects of hBN defects by computationally exploring topological defects, spin qubits, and the microscopic origin of color centers. In our first study, we unravel the intricate relationship between carbon contamination and topological defects in hBN. Carbon not only stabilizes Stone-Wales configurations but also resolves energetic bonds at grain boundaries, leading to preferential accumulation of carbon at grain boundaries. We identify carbon-related defects as potential color centers emitting in the visible range, opening avenues for the creation of visible emitters within hBN.

Our second study focuses on point defect quantum bits in semiconductors, proposing symmetric carbon tetramers in hBN as chemically stable spin qubits for sensing in low dimensions. Through periodic-DFT and quantum chemistry approaches, we predict the electronic, optical, and spin properties of the studied defect. The nitrogen-centered symmetric carbon tetramer exhibits spin state-dependent optical signals with strain-sensitive intersystem crossing rates, showcasing potential advancements in high-resolution, low-dimensional sensing.

The third study delves into the challenge of identifying the microscopic origin of color centers in hBN, particularly the blue emitter. We propose the negatively charged nitrogen split interstitial defect as a plausible model for the blue emitter. A comprehensive theoretical study, addressing the accuracy of first principles methods, reveals the inadequacy of commonly used hybrid exchange-correlation functionals. Fine-tuning the functional, we obtain a zero-phonon photoluminescence energy in the blue range, demonstrating high emission rates and insensitivity to electric field fluctuations.

c-BN epitaxial growth mechanism in ion-beam-assisted MBE

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Cubic boron nitride (c-BN), a metastable phase of BN with sp^3 bonding, has attracted attention as an ultrawide bandgap semiconductor for high-power electron device applications because of the large bandgap energy (6.3 eV) and electrical conduction control [1, 2]. We have achieved epitaxial growth of c-BN layers on diamond substrates by developing a new growth method named ion-beam-assisted MBE [3]. Here, we investigated the influence of the surface orientation on the ion-beam-assisted MBE growth of c-BN layers to gain insight into the growth mechanism.

c-BN layers were grown on diamond (001) and (111) substrates by ion-beam-assisted MBE using electron-beam evaporation of boron and nitrogen radicals (N^*) with a simultaneous supply of Ar^+ . Here, momentum transfer by Ar^+ is inevitable for forming BN sp^3 bonding. The boron flux was 8.4×10^{13} atoms/cm²·s, and the N_2 flow rate for the N^* source was 1–4 sccm.

Figure 1 shows growth rates of c-BN (001) and (111) epitaxial layers as a function of N_2 flow rate. Assuming all supplied B atoms are incorporated into c-BN, the estimated growth rate is 36 nm/hour. However, the obtained ones were less than 26 nm/hour. The reduction in the growth rate is attributed to etching of c-BN and enhanced desorption of surface adatoms by the Ar^+ irradiation. Interestingly, the growth rate of c-BN (001) layers is about twice that of c-BN (111) layers, which means the supplied B atoms are more efficiently incorporated as c-BN on the (001) surface. The surface orientation dependence of the growth rate can be explained by the difference in bonding structures on the surfaces, as shown in Fig. 2. The number of back bonds on the c-BN (001) and (111) surfaces are two and one, respectively; therefore, on the c-BN (001) surface, adatoms bond strongly with subsurface atoms and are then more efficiently incorporated. Thus, the interplay between the formation of sp^3 bonding via momentum transfer from Ar^+ and desorption of surface adatoms by Ar^+ irradiation plays an important role in c-BN ion-beam-assisted MBE.

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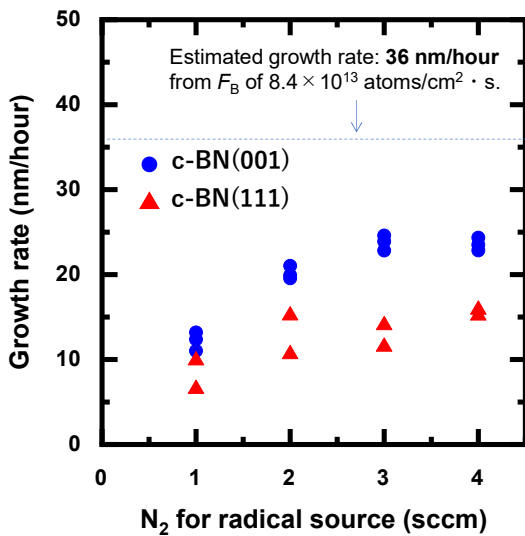


Fig. 1. Growth rate of c-BN (001) and (111) layers as a function of N_2 flow rate.

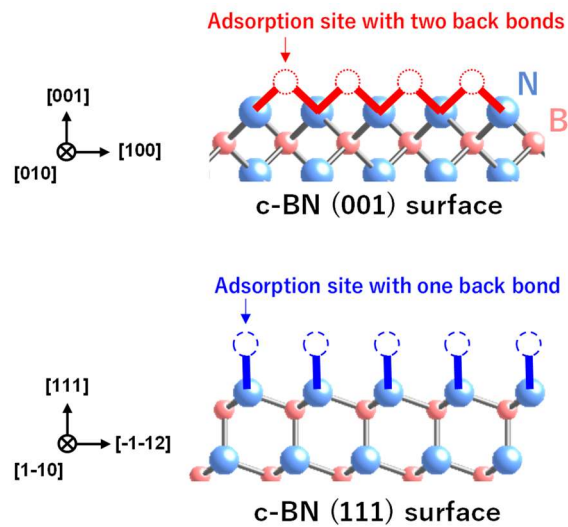


Fig. 2. Atomic arrangement models for c-BN (001) and (111) growth surfaces.

CVD-Grown Hexagonal Boron Nitride for 2D Van der Waals Devices and Quantum Photonics

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Two-dimensional (2D) insulating hexagonal boron nitride (hBN) has been proven to be an important building block for constructing 2D van der Waals (vdW) devices. Recently, hBN has also emerged as an ideal platform for quantum photonic applications, such as hosting single-photon emitters (SPEs) and optically addressable spin defects. For industrial applications, wafer-scale and large-area growth of highly crystalline monolayer and multilayer hBN films is essential. In this regard, hBN films grown by chemical vapor deposition (CVD) is attractive because of their scalability for these applications. Although wafer-scale single-crystal monolayer hBN has been achieved by CVD growth, the effectiveness of using such monolayer hBN as interfacial or encapsulating layers in vdW devices has yet to be demonstrated. The major stumbling block is the inevitable process of transfer from the growth substrate, which can cause organic residues and morphological defects (wrinkles and cracks). The same obstacle is also present for using CVD-grown multilayer hBN films in quantum photonic applications, since the transfer induced defects can cause strong background emissions and degraded SPEs. In this talk, I will present our recent efforts on the CVD-growth of monolayer and multilayer hBN films, as well as their effectiveness for applications in vdW devices and quantum photonics. Top-gate and back-gate transistors based on all CVD-grown MoS₂ with single-crystal monolayer hBN as the interfacial layer will be presented. Considerable improvements in the carrier mobility, dielectric doping and interfacial trap density by the single-crystal monolayer hBN will be discussed. For quantum photonic applications, the creations of SPEs and spin-active boron vacancy defects in CVD-grown multilayer hBN films will be demonstrated. The optical quality of SPEs and electronic structures of the spin-active defects will be presented and discussed.

Optical Crystals of Two-dimensional Rhombohedral Boron Nitride

Kaihui Liu

Peking University

Nonlinear optical crystals play a significant role in advancing laser technology and devices, as well as facilitating the crucial functionalities such as frequency conversion, signal modulation, and parameter amplification. Over the last few decades, the utilization of well-established materials for nonlinear optical crystals like BBO, LiNbO₃, and KBBF has contributed to the fast development of quantum light sources, photonic integrated circuits and ultrafast lasers. The pursuit of suitable nonlinear optical crystals has led the exploration of the potential in two-dimensional (2D) materials, in which boron nitride (BN) is particularly promising due to its high nonlinear susceptibility, broadband transparency, remarkable physicochemical stability, and compatibility with Si-based optical chips. However, the significant challenge of achieving the preparation of single-crystal BN layers is the atomically precise control on domain orientation and interlayer stacking. In this presentation, I will introduce some recent progresses in the growth of large single-crystal BN layers with both in-plane and out-of-plane controls, as well as the development of the twist-phase-matching theory for the design of BN nonlinear optical crystals¹⁻⁴. We believe these findings hold the potential to pave the way for the integration of 2D materials into photonic and optoelectronic applications.

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Self-introduction



Kaihui Liu is currently a professor of Physics at Peking University. His current research interests are the designed growth of 2D single-crystals and the corresponding optical device applications. His main academic achievements include (i) designing the interfacial engineering technique for the universal growth of meter-sized 2D single-crystals, and (ii) fabricating new-concept 2D materials optical fibre and optical crystal devices. In recent years, professor Liu has published more than 60 first or correspondence-authored papers, including Nature (2), other Nature series (21) and PRL (2). He received the National Science Fund of China for Distinguished Young Scholars in 2020, the Xplorer Prize in 2021, and the First prize of Beijing Science and Technology in 2022. Currently, Prof. Liu serves as the dean of the Institute of Condensed Matter and Materials Physics at Peking University and the project leader of China's National Key R&D Program. His research works are selected for 2020 China's major technological progress.

MOVPE growth and applications of layered boron nitride

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Hexagonal boron nitride (h-BN) attracted great interest due to its versatile range of applications. It is clear that the bottleneck for industrial applications of hBN and other 2D materials is the possibility to fabricate high-quality large-area layers.

In the first part of my presentation I will address this issue and show results on the growth of epitaxial h-BN on sapphire by metalorganic vapour-phase epitaxy (MOVPE) [1-4], which is currently regarded as one of the most promising growth techniques. I will show that the growth of a 2D material on a conventional substrate at high temperatures followed by a cool down to room temperature will lead to wrinkle formation [1,5, 6], which can be used to assess the quality of the grown layer.

In the second part, I will focus on the properties of h-BN in general and of our epitaxial h-BN in particular, with a detailed characterization and discussion of the properties.

The last part will be dedicated to applications of our MOVPE-grown h-BN ranging from large-area growth of vdW heterostructures [7], defects and single photon emitters [3, 8, 9], photonic applications like the growth of Bragg mirrors [10] to hydrogen generation and storage applications [1].

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Bound states in the continuum in monolithic van der Waals metasurfaces

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Abstract: We merge bound states in the continuum for strong light-matter interactions and the exceptional properties of van der Waals materials, demonstrating resonances in the visible range in hBN metasurfaces, and self-hybridized exciton-polaritons in WS₂ metasurfaces.

Main Text

Layered van der Waals (vdW) materials, such as hexagonal boron nitride (hBN) and Transition Metal Dichalcogenides (TMDCs), display exceptional optical properties, establishing them as a compelling platform for investigating and controlling nanoscale light-matter interactions. When in their atomically thin monolayer configuration, these materials exhibit appealing characteristics, including strongly bound excitons or optically active spin defects. In contrast, their bulk form displays significant optical anisotropy and high refractive index values exceeding 4, surpassing those of conventional semiconductor materials. This inherent quality makes vdW materials highly attractive for achieving low-loss optical resonances and vertically stacking diverse materials to create innovative all-dielectric nanophotonic structures.

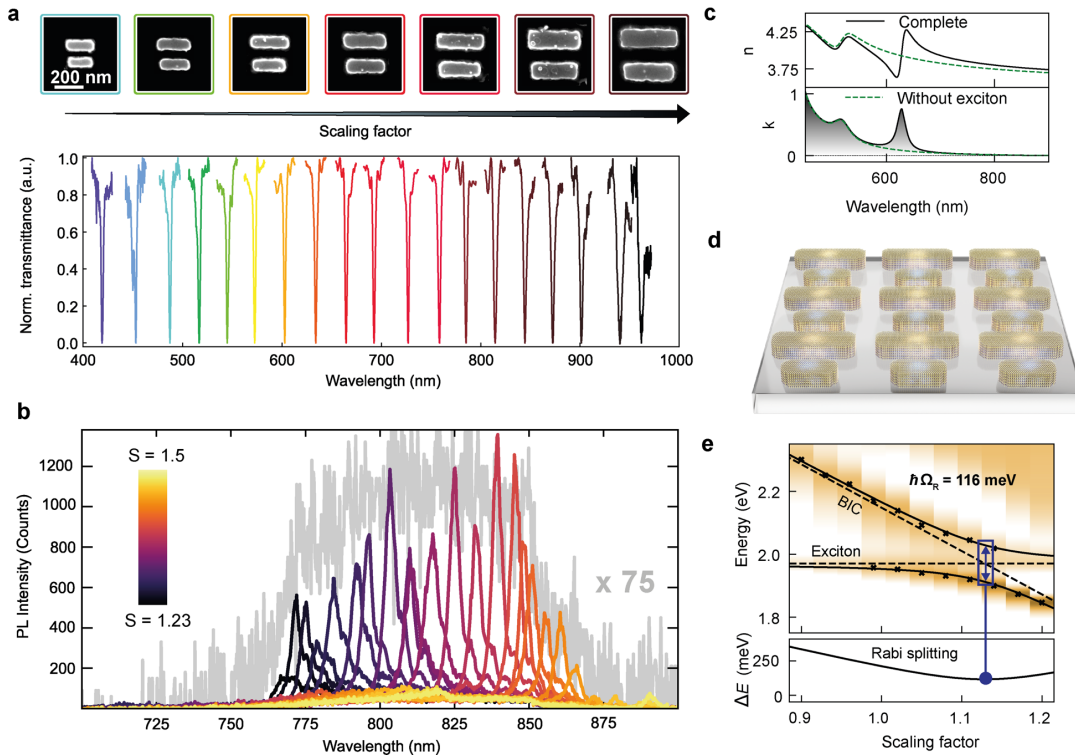


Figure 1 **a** Tuning of the qBIC resonance in hBN metasurfaces across the entire visible range, by scaling the sizes of the metasurface unit cell by a scaling factor (S). **b** PL emission enhancement of hBN spin defect ensemble coupled to qBIC metasurfaces with different scaling factor. In grey, the reference PL spectra from a defect ensemble. **c** Real and imaginary part of WS₂ refractive index. **d** Illustration of a qBIC metasurface in a WS₂ crystal with asymmetric double rod unit cell. **e** Transmittance of WS₂ metasurfaces with different scaling factors, exhibiting the anti-crossing signature of exciton-polaritons, with Rabi energy splitting of 116 meV at ambient conditions.

In our work, we leverage the concept of quasi-bound states in the continuum (qBIC) to generate high quality (Q) factor optical resonances in dielectric metasurfaces composed exclusively of hBN or TMDCs. This monolithic approach achieves optical resonances with Q factors surpassing 10^2 through a two-step fabrication process.

In hBN qBIC metasurfaces, we demonstrate spectral tuning across the entire visible spectrum (Figure 1a), aided by the large transparency window of hBN [1]. Additionally, we prove the enhancement of light-matter coupling with intrinsic negatively charged boron vacancies spin defects in hBN. As shown in Figure 1b, we observe a remarkable increase in photoluminescence intensity and spectral narrowing of defect emissions when coupled to the qBIC resonances. We report a emission funneling in the high Q factor resonance, resulting in a linewidth below 4 nm full width at half-maximum. Furthermore, the qBIC-driven enhancement results in increased spin read-out efficiency for sensing and imaging applications [2].

Finally, our platform presents intriguing possibilities for strong light-matter coupling, exemplified by the distinct anti-crossing behavior between qBIC resonances and intrinsic excitons in monolithic TMDC WS₂ metasurfaces. The self-hybridization of excitons and qBIC resonances results in Rabi splitting values up to 116 meV under ambient conditions. We further show this coupling can be tuned with the geometrical asymmetry of the unit cell, and that is independent of material intrinsic losses [3].

Our findings highlight the potential of combining qBIC photonic metasurfaces with vdW materials, paving the way for hybrid nanophotonic platforms and room temperature polaritonic devices.

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Hexagonal Boron Nitride Nanophotonics: Ultraviolet Transparency, High Refractive Index and Optical Anisotropy

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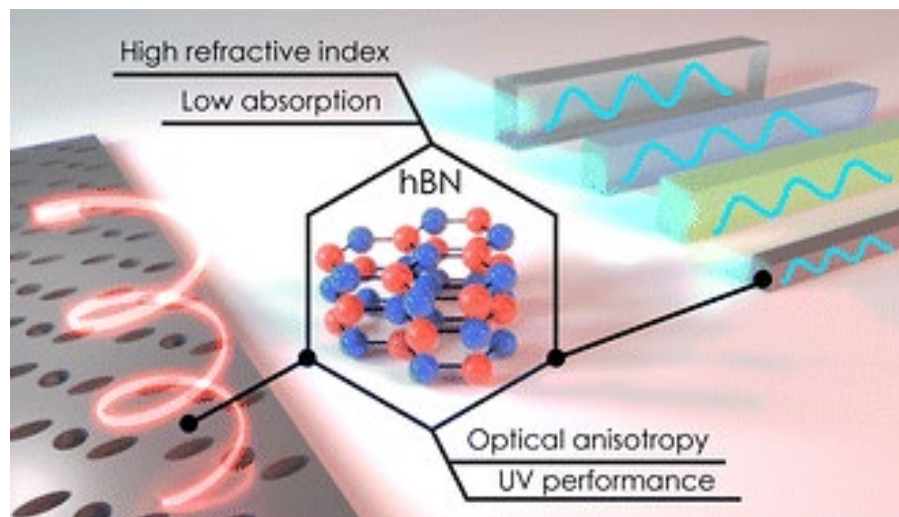
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High refractive index, optical anisotropy, and transparency are the main figures of merit for the materials which define modern nanophotonics. Novel natural materials combining all three properties are in high demand. In this work, we present hBN as a promising material for the visible and ultraviolet range. We determined the exact values of anisotropic dielectric permittivity tensor of hBN in the broad spectral range (250–1700 nm) using cross-validation of far- and near-field techniques, accompanied by first-principle calculations. Our results show high refractive index, transparency over the whole studied spectral range and giant optical anisotropy of $\Delta n \sim 0.7$.

Based on our measurement results, we propose and design novel optical elements: handedness-preserving mirrors and subwavelength waveguides with dimensions of 40 nm operating in the visible and UV range, respectively. Remarkably, our results offer unique opportunity to bridge the size-gap between photonics and electronics.



Interfacial engineering for wafer scale synthesis of multilayer sp²-BN films

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Hexagonal boron nitride (hBN) is a wide band gap layered material, which has been explored as a 2D insulator in „More Moore” ultrascaled devices [1] and as an active medium in „More than Moore” memristor architectures [2]. Achieving consistent device performance is critical to these applications and necessitates hBN of predictable properties on a wafer scale. Interface engineering provides a means for controlling crystal growth, but the processes at the hBN/substrate interface are poorly understood. Here we study sp²-BN film formation on c-plane sapphire in a Close Coupled Showerhead reactor via chemical vapor deposition (CVD) using borazine (B₃N₃H₆), which provides a high yield and carbon-free route to BN synthesis. At growth temperatures of 1350 °C and low BN growth rates, roughening of the underlying substrate is observed (Figure 1a), likely due to competing AlN growth which leads to pinhole formation in subsequent sp²-BN film. Sapphire nitridation indicates active nitrogen species present during borazine pyrolysis playing an important role in sp²-BN film formation [3]. Increasing the BN growth rate promotes layer-by-layer growth up to a critical film thickness, after which 3D particles form, as commonly observed for continuous flow growth of sp²-BN films on sapphire [4]. Here, pinhole-free 3 to 7 nm sp²-BN films with minimal particle coverage (Figure 1b, c) were transferred to Si substrates (Figure 1d, e) for application in resistive-switching devices. We show that interfacial CVD grown graphene protects the sapphire surface, preventing AlN formation during BN growth and facilitating layer-by-layer sp²-BN growth down to 1200 °C. Our findings shed light on mechanisms governing BN film growth on sapphire and show a promising route for hBN/graphene heterostructure integration.

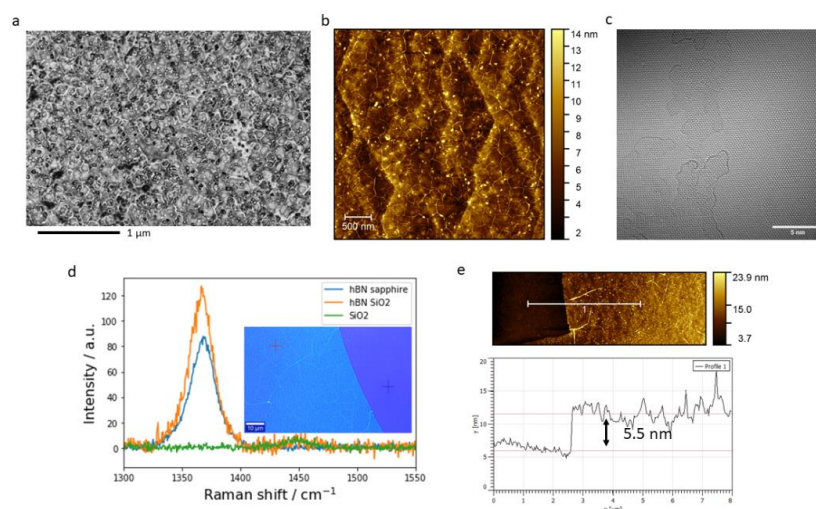


Figure 1. hBN properties; (a) SEM image of AlN formation on sapphire; (b) AFM topography image of multilayer sp²-BN film and (c) corresponding TEM image; (d) Raman spectroscopy characterization of as-grown material on sapphire and after transfer to SiO₂/Si substrate; corresponding areas of measurement (inset); (e) AFM of transferred sp²-BN film with corresponding height profile.

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Wafer-Scale AA-Stacked Hexagonal Boron Nitride Grown on GaN Substrate

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Hexagonal boron nitride (h-BN) is a two-dimensional layered insulator, drawing significant attention owing to its fascinating properties and promising applications across the fields of photonics, quantum optics, and electronics. The stacking sequence of this layered material is a pivotal factor, determining a variety of h-BN polytypes characterized by distinct physical properties. Almost all h-BN layers exhibit the AA'-stacking sequence, representing the most thermodynamically stable structure for sp²-bonded h-BN. Here, we demonstrate the scalable synthesis of h-BN featuring unprecedented AA stacking, where atomic monolayers align along the c-axis without any translation or rotation. This presumably unstable h-BN polytype is achieved through epitaxial growth on a 2-inch single crystalline gallium nitride (GaN) wafer, employing a metal-organic chemical vapor deposition (MOCVD) technique. The formation of AA-stacked multilayer h-BN is evidenced by combining fifth-order aberration-corrected scanning transmission electron microscopy with optical experiments including second harmonic generation, second-order resonant Raman scattering, and photoluminescence spectroscopy in the deep-ultraviolet range. These findings are supported by comprehensive interpretations derived from density-functional theory calculations. The mechanism underlying the formation of this unprecedented atomic stacking is explored through structural and electrical characterizations, along with theoretical modeling, revealing the crucial role of electron doping in stabilizing the AA stacking. Our findings unveil new perspectives for the scalable synthesis of engineered h-BN polytypes, characterized by unique properties such as large optical nonlinearity and piezoelectricity.

Enhancing Coherence Properties of V_B^- in hBN

Optically active spin defects in van der Waals materials offer a promising platform for advancing quantum technologies. Spin defects in hexagonal boron nitride have attracted significant research interest for their innate ability to integrate with heterogeneous optoelectronic and nanophotonic devices. Focusing on the negatively charged boron vacancy center, we propose and demonstrate two complementary pathways to improving its spin properties for quantum applications.

First, by employing advanced dynamical decoupling sequences that selectively isolate different dephasing sources, we observe more than 5-fold improvement in the measured coherence times. Crucially, we identify that the many-body interaction within the ensemble plays a substantial role in the coherent dynamics, which we utilize as a direct metric for V_B^- concentration.¹

Second, we demonstrate that isotope engineering of hBN as another fundamental solution. We observe substantially narrower and less crowded V_B^- spin transitions, extended coherence time T_2 , and relaxation time T_1 in our isotopically purified $h^{10}B^{15}N$ samples. V_B^- centers samples exhibit a factor of 4 (2) enhancement in DC (AC) magnetic field sensitivity. The individual addressability of the V_B^- hyperfine levels enables the dynamical polarization and coherent control of the three nearest-neighbor ^{15}N nuclear spins.²

Our results provide new insights into the properties of V_B^- which will be important for its future use in quantum applications. The method of isotope engineering is also readily applicable for improving spin qubits in a broad family of van der Waals materials.

¹ Gong, Ruotian, et al. "Coherent dynamics of strongly interacting electronic spin defects in hexagonal boron nitride." *Nature Communications* 14.1 (2023): 3299

² Gong, Ruotian, et al. "Isotope engineering for spin defects in van der Waals materials." *Nature Communications* 15.1 (2024): 104.

The electronic band structure of mono-, bi-, and trilayer h-BN

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Abstract

The strong correlation in 2D materials has been widely studied especially since the superconductivity appear in magic-angle bilayer graphene superlattice. The degrees of freedom in stacking angle in 2D system provides an opportunity for creating artificial superlattice structure, in other words, artificial electronic band structure. Though the complicated artificial structure (large supercell) and many-body effect remain challenging to resolve in nowadays, the artificially controllable structure still holds limitless potential for exploring fundamental science and pushing the cutting-edge technology. The core of the strong correlated 2D systems points to the interlayer coupling. Here we take hexagonal boron nitride (h-BN) as an example, providing a simple physics picture to explain the link between stacking configuration and electronic band structure in mono-, bi-, and trilayer systems. The layer-dependent h-BN ARPES spectra showcase the corresponding π bands number, and the degeneracy or split of π bands at the Brillouin-zone K point are the results of symmetry or symmetry breaking stacking sequence. For monolayer h-BN, the inversion symmetry breaking lattice lead to the energy band gap. For bilayer and trilayer h-BN, the symmetry and symmetry breaking stacking sequence lead to the band degeneracy and split characteristic respectively.

Keywords – hexagonal boron nitride (h-BN), inversion symmetry breaking, band structure

Quantum sensing with single spin defects in boron nitride nanotubes

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Abstract

The recent discovery of spin qubits in hexagonal boron nitride (hBN), a two-dimensional (2D) van der Waals (vdW) material, has opened exciting possibilities for quantum sensing [*Advances in Physics: X*, 8, 2206049 (2023)]. We have demonstrated high-contrast plasmon-enhanced boron vacancy spin defects in hBN for quantum sensing [*Nano Letters*, 21, 7708 (2021)] and achieved optical polarization and coherent control of nuclear spins in hBN at room temperature [*Nature Materials* 21, 1024 (2022); *Nature Communications* 15, 104 (2024)], paving the way for manipulating nuclear spins in vdW materials for quantum information science and technology applications. Recently, we observed single optically active spin defects in boron nitride nanotubes (BNNT), a one-dimensional (1D) vdW material [arXiv:2310.02709 (2023)]. Our findings suggested that these BNNT spin defects possess a spin $S=1/2$ ground state without an intrinsic quantization axis, leading to orientation-independent magnetic field sensing. We have developed a method to deterministically transfer a BNNT onto an atomic force microscope (AFM) cantilever and use it to demonstrate scanning probe magnetometry.

Nitrogen isotopes effects on hBN quantum sensor

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Boron vacancy defect (V_B) in hexagonal boron nitride (hBN) is capable of optically detected magnetic resonance (ODMR) at room temperature [1] and is used as a quantum sensor. Since hBN is a van der Waals material, the quantum sensors can approach the measurement target with nanometer proximity. It is an advantage over nitrogen-vacancy (NV) centers in diamonds, a representative point defect quantum sensor.

The sharper the ODMR spectrum, the better the magnetic field sensitivity; narrower linewidths and higher contrasts are beneficial. The linewidth of a typical NV center is less than 10 MHz. In contrast, the one of a V_B defect extends to more than 100 MHz. The main factor limiting this linewidth is the surrounding nuclear spins. The nuclear spins of the three nearest ^{14}N cause the ODMR spectrum to split into seven parts at 45 MHz intervals. Furthermore, these resonances overlap significantly due to boron nuclear spins. This broadening also reduces the occupancy of states per unit resonance frequency, making it challenging to obtain high contrast.

In this study, we investigated the nitrogen isotope effect on the ODMR spectrum of V_B [2]. ^{15}N , which has only 0.4% of the natural composition ratio, has less spin angular momentum than ^{14}N . It contributes to reducing the number of splits in ODMR spectrum. ^{15}N isotope-enriched hBN was synthesized using a metathesis reaction. The Raman shift of the synthesized hBN was consistent with the change in the reduced mass according to ^{15}N purity. In hBN almost perfectly isotopically enriched to ^{15}N , the spectral splitting due to nearest nitrogen was reduced to four. The increase in sensitivity associated with this isotope effect was estimated to be 1.8-fold. The well-separated resonance signals obtained here are also beneficial in increasing sensitivity when using multi-frequency composite microwave pulse sequences [3].

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Molecular beam epitaxy of carbon-doped hexagonal boron nitride on HOPG: Insights into the atomic structure of single photon emitters

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Recently, there has been a growing interest in the optical properties of hexagonal boron nitride (hBN) for applications in deep-UV photonics and single photon emission. One promising avenue for inducing single photon emission involves carbon-doping of hBN layers.^[1,2] However, while this approach has been shown to be effective, the microscopic structure of the resulting single photon emitters (SPEs) in hBN has yet to be fully characterised. We report the growth of nominally undoped and carbon-doped hBN layers on highly-oriented pyrolytic graphite (HOPG)^[3] by high temperature molecular beam epitaxy (HT-MBE). To gain insight into the nature of carbon-related defects, we have employed low-temperature scanning tunnelling microscopy/spectroscopy (LT-STM/S), synchrotron-radiation photoemission spectroscopy, and near-edge x-ray absorption fine structure (NEXAFS) to study the carbon-doped hBN layers grown on HOPG substrates. We have observed increases of up to ~40% in the number of atomic scale defects in carbon-doped hBN compared to nominally undoped hBN, confirming successful incorporation of carbon in hBN by HT-MBE. High-resolution LT-STM imaging reveals defects consisting of atomically-precise few-atom carbon substitutions (Fig 1a), and STS mapping is used to observe mid-gap features which, we argue, are related to the presence of defects (Fig 1b). Overall, our findings shed light on the microscopic structure of SPEs induced by carbon-doping of hBN layers, which has important implications for the development of new optical devices and technologies.

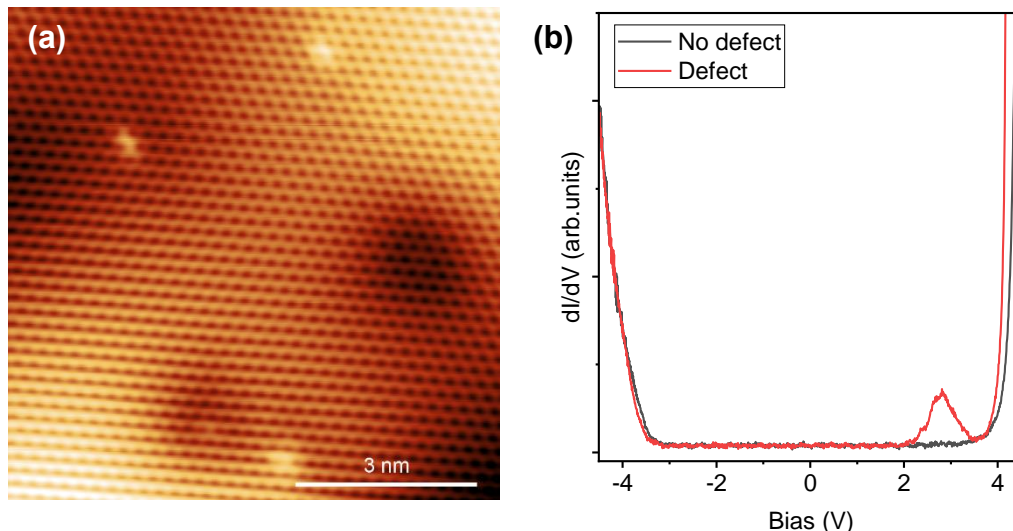


Fig. 1. (a) LT-STM image of carbon defects in bilayer hBN on HOPG (-3.4 V, 20 pA, 4.3 K); (b) STS measurements of pristine (black) and defective (red) in 3L hBN.

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Room temperature electroluminescence from isolated colour centres in van der Waals semiconductors

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Defects in wide bandgap semiconductors have recently emerged as promising candidates for solid-state quantum optical technologies. Electrical excitation of single emitters may pave the way to scalable on-chip devices, and therefore is highly sought after. Here, we demonstrate for the first-time room temperature electroluminescence from isolated colour centres in hexagonal boron nitride (hBN). We harness the van der Waals (vdW) structure of two-dimensional materials, and engineer nanoscale devices comprised of graphene - hBN - graphene tunnel junctions. Under an applied bias, charge carriers are injected into hBN, and result in a localised light emission from the hBN colour centres. Operating at room temperature, our devices exhibit robust, narrowband emission spanning a wide spectral range from visible to near-infrared. Our work marks an important milestone in van der Waals materials and their promising attributes for integrated quantum technologies and on-chip photonic circuits.

Searching for diffuse defects in millimeter-sized h-BN crystals

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Hexagonal boron nitride (hBN) thin film deposition processes can now achieve wafer-scale coverage, yet self-standing hexagonal boron nitride crystals provide exfoliated nanosheets (BNNS) of unrivalled purity and crystal quality which are still preferred for fundamental applications. Defect-free BNNS favour the expression of adjacent 2D materials properties and present interesting intrinsic properties such as hyperbolic phonon polariton (HPP) propagation.

In order to obtain high quality and large size BNNSs, we propose a specific bulk hBN synthesis process at moderate pressure and high temperature. [1,2] The millimetre-sized hBN crystals produced by this method has already demonstrated state-of-the-art crystalline quality and optical properties. [2] Our work aims at understanding hBN crystal growth in order to provide hBN with controlled properties. Indeed, finely controlling doping and/or defect generation is required to tune hBN properties without altering the properties of adjacent 2D materials.

G-FET encapsulated in BNNS exfoliated from our crystals show an electron temperature much higher compared to similar devices made with HPHT hBN while maintaining a high carrier mobility (up to 85 000 cm²/Vs). [3] This high mobility confirms the BNNS crystalline quality whereas the suppression of radiative cooling induced by HPP damping or backscattering is a sign of diffuse defects.

On the basis of these results, we investigate the source of the HPP damping from a synthesis point of view and through optical and structural characterization techniques applied to hBN.

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Optical and spin properties of boron-vacancy centers in few-layer thick hexagonal boron nitride

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Optically-active spin defects in hexagonal boron nitride (hBN) are promising quantum systems for the design of two-dimensional quantum sensing units offering optimal proximity to the sample being probed. In this work, we study the optical and spin properties of boron vacancy centers (V_B) hosted in ultrathin hBN flakes. We first investigate how the photoluminescence signal evolves with the hBN thickness. We observe a non-trivial behavior, which results from thickness-dependent variations of the absorption and the radiation pattern of V_B centers. We then show that the electron spin resonance frequencies of boron vacancy centers (V_B) can be detected optically in the limit of few-atomic-layer thick hBN flakes despite the nanoscale proximity of the crystal surface that often leads to a degradation of the stability of solid-state spin defects. We finally analyze the variations of the electronic spin properties of V_B centres with the hBN thickness with a focus on (i) the zero-field splitting parameters, (ii) the optically-induced spin polarization rate and (iii) the longitudinal spin relaxation time. This work provides important insights into the properties of V_B centres embedded in ultrathin hBN flakes, which are valuable for future developments of foil-based quantum sensing technologies.

Photon statistics analysis of h-BN quantum emitters with pulsed and continuous-wave excitation through Mandel Q

Authors:

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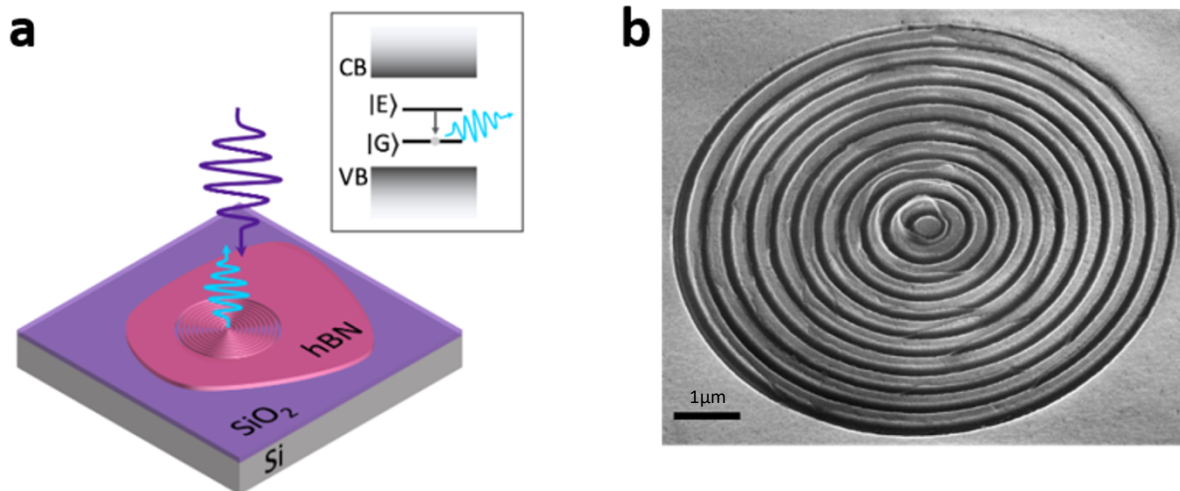
We report on the photon statistics of single photon emitters (SPEs) associated with color centers in hBN using Mandel Q as comparative metric under pulsed and continuous laser irradiation. In this talk, we will present hBN SPEs with statistically significant negative Mandel Q values of -0.002 and -0.0025 under pulsed and continuous-wave laser excitation, respectively. To date, these are the most negative Mandel Q values reported for each type of excitation. These values represent good agreement with expected Mandel Q values accounting for our optical losses. Further, we use an extended version of the Jaynes-Cummings Model to simulate a near-ideal two-level SPE with a Mandel Q of near -1 and demonstrate that including known nonidealities causes the Mandel Q to remain negative but trend towards 0. Under, continuous-wave excitement we define an onset of coherence by varying the time-bin size while minimizing the effect of deadtime. We then discuss the correlation of pump power and blinking with minimum Mandel Q. Finally, we will also establish how Mandel Q can be used to both predict and improve the performance of integrated emitters for quantum applications, using random number generation.

Single photon emitters in hBN are a promising platform for nanophotonic device integration, with many possible applications in quantum photonics. The photon statistics of an ideal emitter would allow for the detection of exactly one photon per time bin, representing a sub-Poissonian distribution. Measuring the Mandel Q value of a source of light quantifies the extent of Poissonian behavior, with negative values representing sub-Poissonian behavior. For a detection of exactly one photon per time bin the Mandel Q would be -1. As such, theory posits Mandel Q = -1 for the ideal SPE. However, due to nonidealities including non-unity quantum efficiencies, collection losses, and detection deadtime, demonstration negative Mandel Q values from SPEs is nontrivial.

Monolithic Integration of Single Quantum Emitters in hBN Bullseye Cavities

Lesley Spencer, Jake Horder, Sejeong Kim, Milos Toth, and Igor Aharonovich*

Quantum emitters hosted as deep level colour centres in hBN are of great importance for quantum photonic applications. In this work we demonstrate enhancement of deterministically placed emitters that emit single photons with a wavelength of 436nm. We couple these single quantum emitters to circular Bragg gratings (CBGs) fabricated monolithically from hBN. We observe a 6-fold increase in the intensity of single photons from an emitter coupled to a CBG compared to an uncoupled emitter, and show exceptional stability at cryogenic temperatures. In addition, we present preliminary etching results beyond the traditional fluorine based reactive ion etching and use electron beam irradiation to create site specific single photon defects. Our work demonstrates the potential of monolithically integrated systems for 14 deterministically placed quantum emitters using a variety of fabrication options.



Ultrafast Thermal Dissipation via Surface Phonon Polaritons
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In recent years, breakthroughs in nanomaterials and radiative engineering have introduced thermal radiation as a promising mechanism to manipulate both the magnitude and direction of heat transfer. However, the peak of blackbody emission at room temperature is in the mid-infrared (MIR) making manipulation of this emission difficult due to the low interactivity of most materials in the region. Conveniently, nanostructuring can give rise to MIR spectral resonances that offer options for potential manipulation of this near-room temperature radiative intensity. These spectral resonances, or "polaritons", arise from strong coupling between vibrations and photons giving the modes dual characteristics of ultrafast speed and high heat capacity. Thus, we can manipulate and facilitate an augmented flow of radiative energy from generic slow-moving heat carriers into specific ultrafast spectral modes and vice versa. One proposed way to direct the radiative energy is by utilizing optical phonons, to couple directly with near-field radiation emitting from a nearby thermal source. As the first identified natural hyperbolic material, hexagonal boron nitride (hBN) supports hyperbolic phonon polaritons at MIR frequencies positioning itself perfectly as a near-field radiative energy sink. Another possibility for this capture relies on capitalizing on the hybridization of resonant modes in plasmonic materials, such as highly doped cadmium oxide (CdO) superlattices that support tunable hyperbolic plasmonic dispersions in the MIR via epsilon-near-zero resonances, while silicon carbide nanowires provide evidence of the dramatic role that phonon polaritons can provide in enhancing thermal conductivity under steady state conditions.

In this work, we report on experimental investigations into the thermal interactions between gold radiating pads and hyperbolic modes within layered hyperbolic metamaterials (HMM) and flakes of isotopically enriched hBN. Our approach involves employing a unique ultrafast pump-probe spectroscopy technique that enables spectral resolution of vibrational modes with sub-picosecond temporal resolution. This technique relies on a frequency-tunable mid-infrared probe pulse, allowing us to directly interface with polaritonic heat carriers, thereby providing a direct assessment of their temporal propagation within nanoscale material systems. By utilizing this method, we investigate the ultrafast thermally induced near-field coupling of hyperbolic plasmon and phonon polariton modes via far-field sensing. Furthermore, we explore the spatial extent and efficiency of direct coupling between thermally excited polaritonic modes and spectrally resonant heat sinks. We thereby demonstrate and quantify the capability of tuning the near-field radiative heat transfer at ultrafast speeds. Our results provide a new path for novel designs of polaritonic devices with far-reaching applications at the intersection of thermal transport and photonics where structure and vibrational selection rules allow for the transduction of thermal energy at speeds faster than diffusion, leveraging this heat flux as a source for photonic communication as well as offering a novel approach to thermal management.

Spin properties of visible emitters in hBN and their applications

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Optically addressable spin defects in hexagonal boron nitride (hBN) have recently begun to be recognised as potentially useful systems for quantum technology applications, with unique attributes afforded by the material's layered van der Waals structure [1]. Recent demonstrations of quantum sensing with the boron-vacancy centre have established the potential usefulness of hBN based sensors in a selection of applications [2, 3]. The visible class of emitters, which have recently been observed by a number of groups offer a potential alternative to the boron-vacancy centre, though their structure is yet to be properly identified. Here we present substantial evidence leading towards uncovering the identity of these emitters by studying ensembles of them in a number of hBN samples. First, conclusive evidence identifying the spin-1/2 nature is presented [4], and is attributed to a weakly coupled spin pair system which we are able to construct a kinetic model for, and verify experimentally. Using insights from the kinetic model we are able to accurately measure the spin relaxation and coherence times. We then demonstrate the sensing capabilities of the visible emitters by imaging a 2D magnetic material, using the isotropy inherent to a spin-1/2 system to measure the target materials anisotropic susceptibility. Finally, we demonstrate how visible emitters are able to coexist with the boron-vacancy centre in the same sample and identify a coupling between them through cross-relaxation. Our results provide new and substantial insights into the visible emitters, laying the ground work for being able to properly identify them in the future. Furthermore, we also realise potential applications, guiding the future of quantum sensors in 2D materials.

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Optical Readout of Redox Reaction via hBN Surface Emitters

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In this work, we leverage a single molecule microscopy approach combined with pristine hexagonal boron nitride (hBN) to develop a nanoscale sensor that can optically track redox reactions in organic solvents. Through a proposed sensing scheme based on redox-active species interaction with fluorescent emitters at the surface of hBN, we observe a linear decrease in the number of emitters while increasing positive voltage, decreasing by more than 50% at +0.75V vs Ag/AgCl in methanol. We also calculate consistent Tafel slopes between potentiostat-reported current and optically read emitter decay kinetics, with behavior characteristic of electrode reaction kinetics vs overpotentials. We also draw on the spectral capabilities of our spectral SMLM to monitor the fluorescent species consistent identity. Finally, we enable time-resolved measurements of redox groups at a micrometer-length scale with nanometer-scale depth and precision capabilities. Considering the scalability and versatile defect engineering options provided by 2D materials, the interactions can be diversified to optically detect a variety of interacting species with the unprecedented localization precision of atomic reaction centers.

Chemical intercalation, exfoliation, and functionalization of hBN materials

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Abstract

Introducing alkali metal atoms between the carbon layers of graphite to form graphite intercalation compounds (GICs), can tune the interlayer spacing and charge the graphite host through a variety of electronic ground states.¹ Hexagonal boron nitride (hBN) is another prototypical layered material and possesses a hexagonal network consisting of B and N atoms. Because of its structural similarity to graphite, there has been several theoretical calculations predicting host-guest redox-driven hBN intercalation compounds.² Based on the analogy with GICs, the hybrid structure of hBN and alkali metal atoms that are intercalated into the interlayer spacing may also exhibit interesting electronic properties and utility as a raw material for 2D materials with unprecedented physicochemical properties.³ However, the intercalation of alkali metal in hBN has proven to be considerably more difficult than graphite. Here, we will present our latest experimental data, showing that potassium metal can be intercalated into hBN layers.⁴ The K-intercalated hBN material and films exhibits unprecedented electronic and energy storage properties.⁵

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Hexagonal Boron Nitride Memristors and RF Switches

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Hexagonal boron nitride (hBN) possesses a substantial bandgap, elevated phonon energies, and a remarkably smooth surface devoid of dangling bonds. This has led to its widespread use as a dielectric within two-dimensional heterostructures, as well as in the construction of two-dimensional transistors and optoelectronic devices. Here, we introduce the applicability of hBN in crafting atomic scale memristors and RF switches tailored for high-frequency communication systems. Our approach takes advantage of the non-volatile resistive switching characteristics that exist in atomically thin hBN.

The hBN memristors feature the forming-free switching in both unipolar and bipolar operations, with large on/off ratio ($> 10^7$), and fast switching speed (< 15 ns) with pulse operation. Simulation results using the ab-initio approach indicate that metal ion substitution into hBN vacancies during electrical switching is a likely mechanism.

The hBN RF switches exhibit a cutoff-frequency figure of merit reaching approximately 129 THz, characterized by a minimal insertion loss (≤ 0.5 dB) and substantial isolation (≥ 10 dB) across the frequency range of 0.1 to 200 GHz. Additionally, the switches showcase a remarkable capacity for high-power handling (around 20 dBm) and nanosecond switching speeds, orders of magnitude more energy efficient DC power consumption than other emerging solid-state switches. The switches also show the potential of the hBN in a communication system with approximately 10 Gbit s⁻¹ data transmission rate at 100 GHz with a low bit error rate.

Electron Beam Restructuring of Quantum Emitters in Hexagonal Boron Nitride

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Hexagonal boron nitride (hBN) hosts a vast library of single photon emitters (SPE), with the B-centre defect garnering particular interest due to its narrow zero phonon line (ZPL) at 436nm and the ability to generate the emitter by site-specific electron beam irradiation [1]. These characteristics make it appealing for integrated on-chip quantum photonics due to ease of incorporation in devices such as photonic crystal cavities [2].

It's known that the emergence of the B-centre is significantly more probable in the presence of the 305nm UV defect in carbon-doped hexagonal boron nitride (hBN) [3]. However, beyond this observation, the fundamental mechanism underlying the generation of the B-centre remains inadequately understood [3]. Here, we use in-situ time-resolved cathodoluminescence (CL) spectroscopy to investigate the kinetics of B-centre generation in hBN by an electron beam.

We demonstrate that: (i) the formation of B-centres coincides with the simultaneous quenching of the carbon-related UV defect in hBN (Figure 1), (ii) electromigration, involving mass transport of defects within the hBN lattice, is a significant factor in the electron beam-induced restructuring of both defects (Figure 2), and (iii) the introduction of water vapor during electron beam irradiation effectively mitigates electron beam-induced carbon deposition effects (Figure 3).

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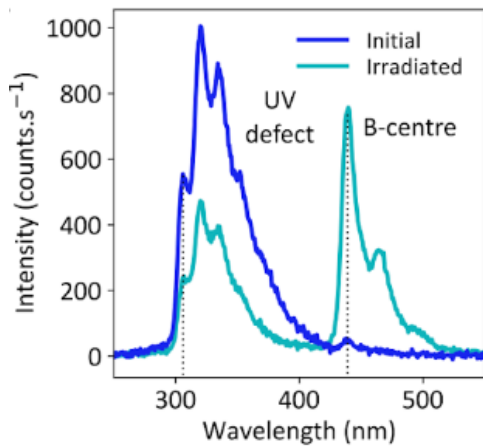


Figure 1 - CL spectra showing the UV and B-centre emissions at the start and end of a 300 s electron beam irradiation treatment. The UV and B-centre ZPLs are indicated by vertical dashed lines. The UV and B-centre emission intensities decrease and increase, respectively, as a result of the electron irradiation. The spectra were acquired using an integration time of 2 s. Electron beam energy = 5 keV, beam current = 1.6 nA.

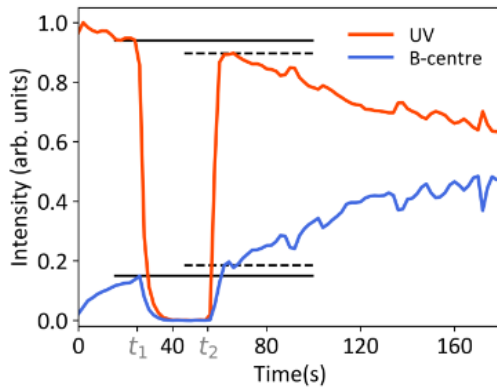


Figure 2 - Role of the electric field ξ in CL kinetics. UV and B-centre CL intensity measured as a function of electron beam irradiation time. The irradiation was interrupted at time t_1 for 30 s and resumed at t_2 . The solid and dashed black lines show the CL intensities at t_1 and t_2 , respectively. The intensities continue to change while the irradiation is interrupted, because ξ does not dissipate instantaneously when the irradiation is interrupted.

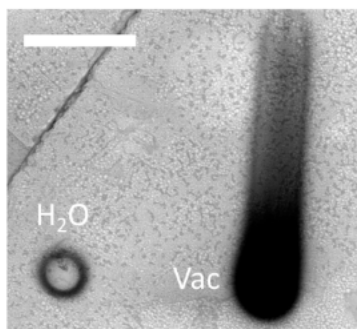


Figure 3 - Water mediated stabilisation of hBN. (a) Two 300 s irradiation spots undertaken in high vacuum (Vac) and with the addition of water via a GIS (H_2O). The scale bar is 5 μm .

Solid states quantum emitters in wide band gap materials for quantum technology applications

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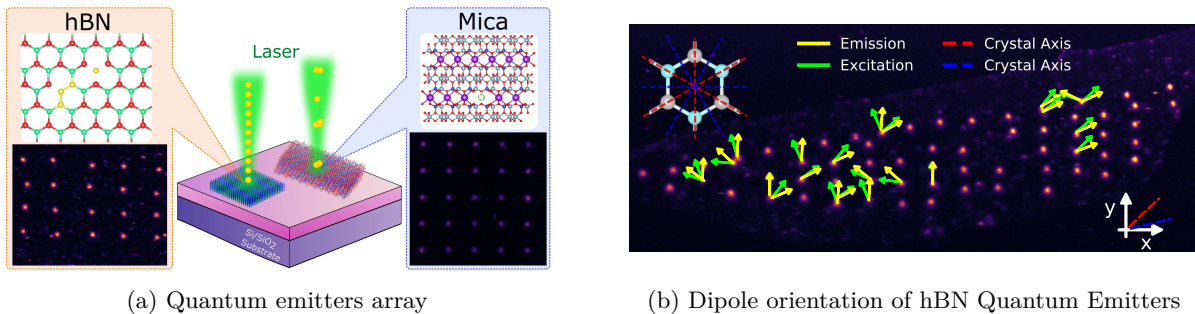
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Quantum emitters in solid-state crystals have gained attention with the recent advancement of quantum technology applications. In particular, fluorescent defects in wide band gap materials stand out due to their room temperature operation and high luminosity. However, the atomic structure of these fluorescent defects is still not well understood yet. Here, we present the generation and investigation of quantum emitters in wide-bandgap materials, including multi-layer hBN and muscovite mica. To complement our experimental observations, we also provided density functional theory calculations. We used localized electron beam irradiation using a scanning electron microscope to induce the defect-based emitters, which could rely on activating pre-existing defects by charge state manipulation. While our localized irradiation produces single photon emitters at 575 nm with high purity in hBN, it lacks to generate bright single emitters in muscovite mica. The polarization-resolved measurements of hBN emitters present a strong correlation of emission and excitation dipole axis with the crystallographic axis of hBN, as confirmed with second-harmonic generation. This correlation of the dipole axes of the emitter with the crystallographic axis provides an important step toward the identification of emitters and their formation process. Additionally, we explore temporal polarization dynamics, uncovering a mechanism that governs the time-dependent polarization visibility and dipole orientation of color centers in hBN and diamond. This is crucial for achieving a lower quantum bit error rate in quantum key distribution. Our further investigation involves the integration of hBN emitters with a nanophotonics platform to develop on-chip quantum light sources for quantum technology applications such as quantum memory and quantum key distribution.



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Thermal property and applications of boron nitride nanosheets

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Heat dissipation is becoming increasingly critical, particularly in modern miniaturized devices. Boron nitride (BN) nanosheets show promise for heat dissipation due to their high thermal conductivity and electrical insulation properties. Here, we present the intrinsic thermal conductivity of atomically thin BN and explore thickness and isotope effects on heat transport (1-2). To enhance the dispersibility of BN nanosheets for advanced composite fabrication, we introduce a simple yet efficient mechanochemical exfoliation technique, yielding functionalized BN nanosheets highly dispersible in both water and organic solvents (3). Leveraging these high-quality BN nanosheet dispersions as precursors, we fabricate BN spheres with high, isotropic thermal conductivity. These spheres substantially enhance the thermal conductivity of poly(vinyl alcohol) by approximately 3700%, addressing the challenge of highly anisotropic thermal conductivity in BN-reinforced polymers (4). Our study not only offers insights into heat transport and phonon scattering mechanisms but also provides a practical pathway for fabricating BN-enhanced thermal interface materials with high isotropic thermal conductivity. These materials hold promise for efficient heat dissipation in cutting-edge advanced electronics and related applications.

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What is the nature of the UV color center emitting at 300 nm in hexagonal boron nitride ?

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The intrinsic optical response of hexagonal boron nitride (hBN) lies in the UV-C spectral range, at wavelengths between 200 and 215 nm close to the ~6 eV band-edge of this ultrawide-bandgap material. The extrinsic optical response due to point defects features well-defined optical lines within the bandgap, particularly in the UV-B domain around 300 nm. The recombination of the well-known “4 eV defect” is ubiquitous in hBN, but its origin is the subject of an intense debate motivating many experimental and theoretical studies [1-5]. Carbon is suspected to be involved in the defect structure [2] but contradictory results in the literature [2-4] cast doubts about its exact role for the formation of this UV-color center.

We will present our recent results by photoluminescence spectroscopy in hBN crystals of controlled isotopic content and polytypic phase. By careful study of the spectral position of the zero-phonon line and phonon side-bands, we provide novel fingerprints for the identification of this point defect and we discuss its possible origin in the light of recent advanced DFT calculations [5].

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Single photon emitters created by intentional carbon doping of hexagonal boron nitride grown on sapphire by high-temperature molecular beam epitaxy.

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In recent years single photon emitters (SPEs) in hexagonal boron nitride (hBN) have been extensively studied using confocal microscopy. Fluorescent spectra using a 532 nm excitation wavelength often show a zero-phonon line at emission wavelengths ~ 575 nm with a phonon sideband at ~ 625 nm; it has been suggested that these bright SPEs are associated with carbon impurities in the layers [1]. Here we report the optical characterisation of hBN grown on sapphire substrates by high-temperature molecular beam epitaxy (HT-MBE) [2]. Some of the samples were grown without intentional carbon doping whilst others were grown in the presence of a controllable flux of carbon. Variable angle spectroscopic ellipsometry (VASE) and X-ray photoelectron spectroscopy (XPS) have been used to determine the thickness of the layers (1 – 10 nm) and the optical constants show defect bands at ~ 4.3 eV and ~ 5.4 eV for the samples grown with the highest carbon fluxes these have previously been associated with carbon impurities [3,4]. Low temperature deep UV photoluminescence measurements ($T = 8$ K, excitation wavelength = 195 nm) of the HT-MBE grown layers show features that may be related to rhombohedral stacking in hBN [5], but do not show strong systemic changes with the carbon flux during growth. Confocal microscopy carried out at room

temperature (532 nm excitation wavelength) show bright single photon emitters with similar fluorescent spectra to those reported in the literature. As the carbon doping is increased the areal density of the SPEs increases until at the highest carbon doping a homogeneous fluorescent response is observed (fig. 1). In summary, hBN can be intentionally doped with carbon during growth using HT-MBE. The fluorescent spectrum of carbon-doped boron nitride is similar to previously reported spectra of SPEs in hBN, adding to the evidence that these emitters are related to carbon impurities in the hBN.

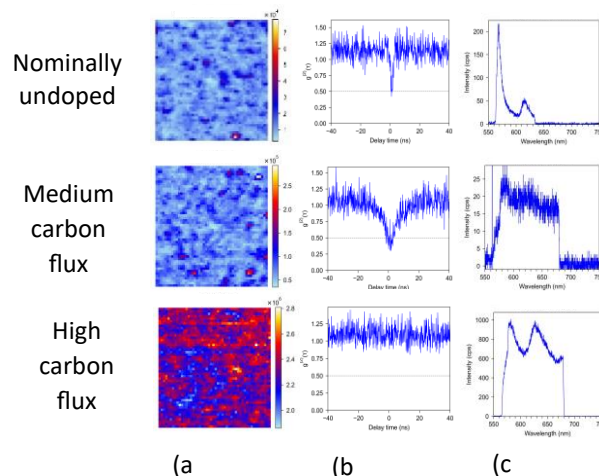


Figure 1a) Confocal fluorescence maps ($10 \mu\text{m} \times 10 \mu\text{m}$) b) Hanbury-Brown-Twiss correlation measurements c) Filtered fluorescence spectra of HT-MBE grown hBN layers on sapphire with different carbon fluxes during growth.

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QUICK³ – Towards satellite-based quantum communication, and fundamental physics tests in microgravity

Abstract by Kabilan Sripathy, FSU Jena

In this second quantum revolution, the 2D material hexagonal boron nitride (hBN) has emerged as a front-runner for substrate materials, largely due to its ability to host compact color centers capable of producing high-purity, tunable single photons at room temperature. One of the most promising applications of these hBN emitters is in the development of quantum key distribution (QKD). With the aim of intercontinental QKD, a crucial step in this endeavor is the testing of these emitters in space.

This is the goal of our current flagship project, QUICK³. In this partnership with FBH Berlin and TU Berlin, along with associate partners at CNR-IFN, NUS and UOC, we are building a 3U CubeSat to contain a hBN emitter integrated into an etched waveguide circuit. The hBN emitter centered at 780nm and filtered through fiber Bragg coatings, emits directly into the laser-etched circuit, which can be tuned using thermal phases shifters. The circuit can then be aligned to perform a coincidence $g^{(2)}$ measurement, thus we can confirm the purity of these single photons low Earth orbit (LEO) environment can match identical ground experiments. In addition, the waveguide circuit has been fabricated to take the form of three parallel Mach-Zehnder interferometers. Consequently, once our CubeSat is in orbit, we will also be able to test for the possible existence of third order interference, and thus deviation from the Born rule in a microgravity environment. While such deviations are speculative, there exist theories (particularly within the framework of pilot wave theory) suggesting their existence.

The flight model of the 3U CubeSat, destined for a sun-synchronous 500 km LEO in 2024 is currently under construction across the collaborating groups. The status of which is reported in detail in our recent publication [1]. Space qualified components are being tested, whilst preliminary experiments have been conducted using engineering models of the waveguide circuit. With the outsourced components coming in, waveguide etched, emitters ready, and novel transfer technique proven, the time for system level tests with the final payload is approaching. With diligence and dedication, QUICK³ will demonstrate the first true single photon source in space.

[1] - N. Ahmadi et.al. | Adv Quantum Technol, 2024, 2300343

Systematically creating boron vacancies in bulk exfoliated hexagonal boron nitride flakes using focused ion beam

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The hexagonal boron-nitride (hBN) is a wide band-gap insulator, which has become an essential ingredient in stacked van der Waals (vdW) hetero structures. Defects in hexagonal boron-nitride (hBN) have been studied using electronic transport- where they are identified as nanoscale quantum dots, sensitive to the spectra and compressibility of nearby samples. They have also been extensively studied by using optical fluorescence. Color centers in hBN, specifically boron vacancies (V_B^- s), are stable spin defects in the hBN lattice with similar traits to the well-known nitrogen vacancies (NV^-) in diamond and have comparable applications.

In this work [1], we focus on generating V_B^- defects in bulk exfoliated hBN using ion implantation, and specifically using a focused ion beam (FIB). Enabling detailed and systematic studies of the implantation parameters, comparing both different parameters on the same flake and different flakes. We focus on sample thicknesses in the range of 50–200nm, which is of interest due to the significant signal obtained above 50 nm, and the advantages of the vdW nature of the material below 200 nm (above which bulk-like behavior begins to emerge).

We present the successful creation of V_B^- s using 12 keV FIB ion implantation, with both nitrogen and oxygen ion beams. The V_B^- defects were characterized for their optical and quantum properties, namely using spectral measurements and ODMR techniques. Our results show that both oxygen and nitrogen ions at 12keV can efficiently create the V_B^- s in hBN flakes of various thicknesses. We find that the cleaning and preparation process before the FIB implantation is crucial for robust creation of V_B^- defects in hBN, and more importantly, we find that the hBN flake thickness plays a key role in the success of the defect creation process and in the achievable V_B^- yield.

Spin-1 Quantum Sensors in hBN: Intersystem Crossing Relaxation of the Metastable State and Irradiation Protocol

The wide band gap material hBN is host to various optically active defects. One of them is the negatively charged boron vacancy V_B^- , which exhibits photoluminescence (PL) around 800 nm under optical excitation. Interestingly, this defect also forms a ground-state spin triplet system that can be efficiently polarized by optical excitation. Spin polarization can be manipulated by applying resonant microwaves at ~ 3.5 GHz. The current spin state can be read-out optically due to spin-dependent change in the PL. This method is referred to as optically detected magnetic resonance (ODMR).

Environmental conditions like temperature, pressure or magnetic fields shift the resonance frequency of the defect center and render the system as a sensor for these parameters. Here, we studied the influence of fluence-variation of nitrogen ions for defect creation and found strong influence on the lifetimes of the system and therefore sensing performance. We investigated the spin-lattice relaxation T_1 as well as the attenuation of Rabi oscillations T_ρ . These results can therefore be very useful as a guide for sample preparation.

Furthermore, we investigated the relaxation dynamics in the excited states in the optical cycle under pulsed laser excitation using transient PL. The focus here was on the metastable state (MS), i.e., the state in which electrons are shelved when the system is optically pumped. This state is crucial for initialization of the triplet ground-state. By varying the time between two consecutive laser pulses we observe significant changes in time-resolved PL from which we derived a relaxation constant of approx. 24 ns at room temperature for the MS. We conclude that accounting for this relaxation significantly increases the effectiveness of the microwave manipulation and thus the sensitivity of the sensor.

Conductivity induced by post growth annealing of boron nitride grown by MOVPE

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Hexagonal boron nitride (hBN) holds significant potential for optoelectronic applications in the deep ultraviolet (DUV) spectral range. A major obstacle in realizing hBN-based devices is the lack of efficient n-type and p-type doping, which would substantially lower the naturally high electrical resistivity of the material. This study explores the impact of post-growth annealing in a carbon-rich environment on the electrical and structural properties of hBN samples grown using Metal-Organic Vapor Phase Epitaxy (MOVPE) [1-3]. The growth temperatures ranged from 1095°C to 1285°C. Following the growth process, annealing was performed at 1400°C in a nitrogen atmosphere for 10 minutes. Resistivity measurements performed before and after the annealing clearly indicate a decrease in the resistivity of our hBN layers, as depicted in Figure 1a. Photoluminescence (PL) and absorption measurements will be presented to show changes in the optical properties introduced by the annealing. To determine the structure of the samples, additional Raman measurements were performed. Interestingly, the Raman spectra of the annealed samples showed strong satellite structures below and above the E_{2g} mode, as presented in Figure 1b. Additionally, the PL background significantly decreases for the annealed sample. The observed changes in the electrical and structural properties of the annealed hBN samples may be attributed to a reaction mechanism wherein annealing in a carbon-rich environment facilitates the removal of hydrogen from boron vacancies within the material and the subsequent incorporation of carbon atoms into these vacancies [4]. The possible tuning of the carrier concentration by dedicated annealing processes is technologically very attractive and may guide the way for future applications of boron nitride.

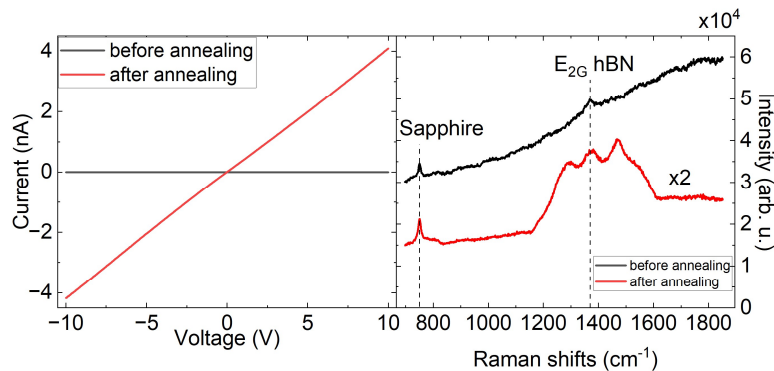


Figure 1. a) IV measurements on both annealed and non-annealed samples using two-point tungsten probes positioned approximately 1 mm apart. b) Raman spectra of the sample before and after annealing, displaying satellite peaks both below and above the main Raman peak of hBN. The spectra for the annealed sample were multiplied by a factor of 2 to compensate for a significant drop in the background PL.

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hBN-Enabled Flexible GaN Photodetector

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Ultraviolet (UV) photodetectors capable of detecting wavelength in the range of 200 to 400 nm have significant applications in the field of environmental monitoring, civil as well as military applications. For instance, such UV photodetectors can warn of harmful UV exposure under the sun in our daily lives. GaN with a direct bandgap of 3.4 eV is a potential candidate for UV photodetector. Herein, we report on GaN-based UV photodetectors, that can be easily delaminated using a scotch tape from the sapphire substrate. They are fabricated and transferred onto another flexible substrate for applications in wearable and flexible electronics.

Our technology is based on first growing a hexagonal-BN film on the sapphire substrate followed by growing the GaN device layer. Due to the 2D properties of hexagonal-BN film, a weak van der Waals force exists at its interface with the GaN layer, which enables easy delamination of the device layer.

Fig. 1(a) shows the fabricated GaN photodetectors in both the photoconductive (PC) as well as metal-semiconductor-metal (MSM) architectures on sapphire substrate. Fig. 1(b) shows the devices after being transferred to a flexible substrate. Fig. 1(c) shows the IV characteristics of the photodetectors. The PC detector shows an order of magnitude enhancement in current from 1.07×10^{-3} to 1.19×10^{-2} A under AM 1.5G illumination, while the MSM detector shows over two orders of magnitude enhancement from 8.07×10^{-8} to 2.28×10^{-5} A at a bias of 3 V. The relatively low operating voltage of 3 V is expected to enable low-power-consuming wearable and flexible UV photodetectors, whilst the operational photocurrent range (mA and μ A) of both detector architectures allows broad compatibility with a range of electronic circuits.

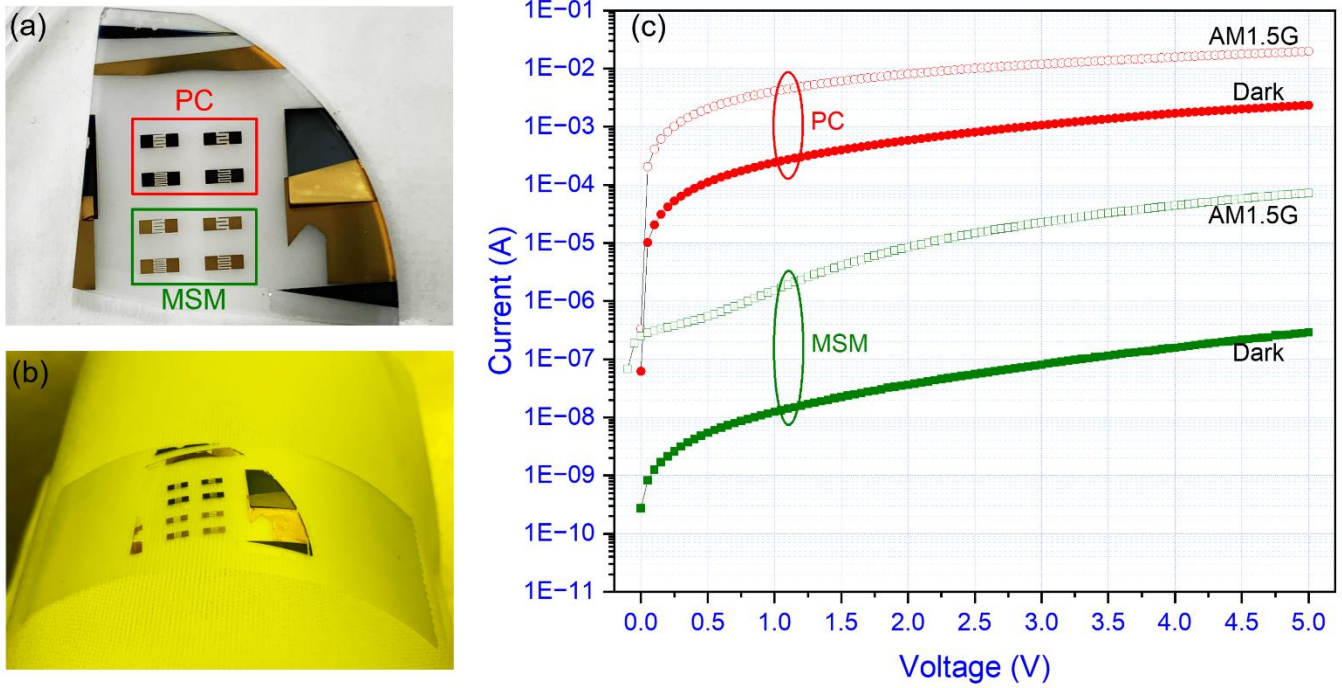


Fig. 1. (a) Photoconductive (PC) and metal-semiconductor-metal (MSM) GaN photodetectors on a sapphire substrate. (b) The photodetectors after transferred to a flexible substrate using a scotch tape. (c) Current-voltage characteristics of the PC and MSM photodetectors under dark and AM1.5G illumination.

Carbon Migration and Single Photon Emission in Electron Irradiated Hexagonal Boron Nitride Flakes

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Electron beam induced defects in hexagonal Boron Nitride exhibit luminescent properties^{1,2}. Various studies have attributed carbon substitution to be the cause of these emission³⁻⁵. In this work we propose high carbon creation at the irradiation site and gradual decrease in carbon concentration away from it. Single photon emitter is observed at the location of lowest carbon.

Center of h-BN flake (Spot 1) was irradiated using 5 keV electron beam in an FESEM system and subsequently annealed at 850 degrees Celsius under continuous argon flow. Optical spectrum obtained from spot 1, 2 and 3 are shown in figure 1b below. A gradual reduction in carbon concentration is eminent from decrease in intensity of 1614 cm⁻¹ G peak and 2700 cm⁻¹ 2D peak. A sharp ZPL at 577 nm (figure b panel 3) was observed at distance of few microns (spot 3) from the electron irradiated area. Antibunching measurement for this emitter (inset of panel 3 figure 1b) gave a $g^2(0)$ of 0.19 indicating presence of single photon. Based on our observations we suggest outward migration of carbon from the irradiated region and appearance of stable emitter at the site of lowest carbon presence. A possible explanation for this behavior is the post irradiation annealing treatment which provides sufficient kinetic energies to the defect complexes and hence promote their movement away from the high carbon concentration sites.

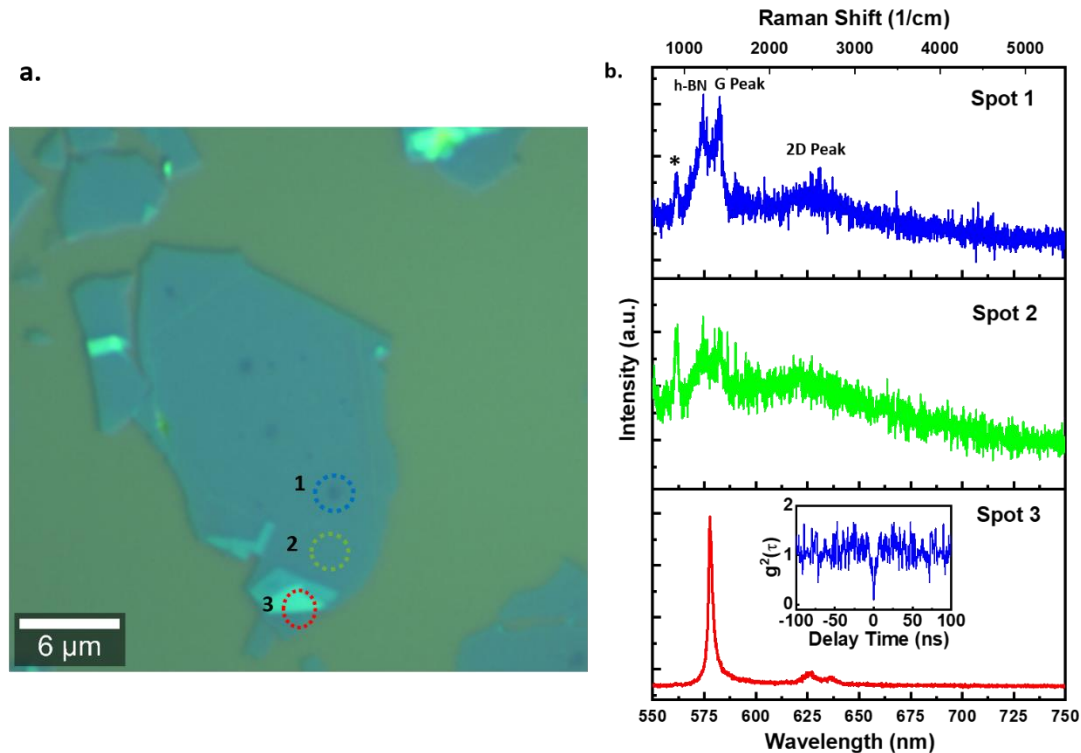


Figure 1.a. Optical Image of the irradiated flake. Encircled region marked 1 is the irradiation spot. b. Optical spectra obtained from regions spot 1, 2 and 3. Decrease in carbon raman peak and observation of Single photon emitter occurs as we move from spot 1 to spot 3. Spot 3 (last panel) spectrum features a sharp ZPL peak at 577 nm with a $g^2(0)$ of 0.19 indicating presence of single photon. * Represent silicon second order raman peak.

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Polytype Identification in MOVPE Grown sp^2 -BN Using Ultraviolet Defect Photoluminescence

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Boron nitride is the only III-nitride which naturally crystalizes in a sp^2 -bonded, layered crystal structure. Within such crystals layers can be stacked in different ways leading to polytypism in the material. The most common polytype is hexagonal boron nitride (hBN) with AA' stacking in which subsequent layers are twisted by 60° with respect to each other. Other observed sp^2 -BN stackings are AB (bBN) and ABC (rBN). Due to the in-plane shift of the subsequent layers, those polytypes exhibit different symmetry showing characteristic additional properties such as second harmonic generation, piezo- and pyroelectric effects. [1] Therefore, for different types of applications, sp^2 -BN with appropriate stacking sequence is required and the identification of the appropriate polytype, as well as the ability to intentionally grow a given sp^2 -BN polytype, becomes an important challenge.

In this work we present a method for polytype identification of epitaxial boron nitride grown by Metal Organic Vapor Phase Epitaxy (MOVPE) [2] using defect photoluminescence in the UV range. There are many hypotheses about the origin of a characteristic, sharp-line emission at about 300 nm [3]. We show that the most probable candidates are specific carbon complexes, which are sensitive to different stacking sequences. Three peaks at 4.10 eV (302 nm), 4.14 eV (299 nm) and 4.16 eV (298 nm) can be used to distinguish the major different polytypes. Our findings are supported by theoretical calculations, X-Ray Diffraction, Transmission Electron Microscopy and Piezoresponse Force Microscopy. We also show that with MOVPE we can influence the growth of different polytypes on the wafer scale, which opens up large possibilities for future applications.

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Boron nitride nanosheet aggregates for enhanced acoustic energy harvesting

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While often perceived as a nuisance, noise represents a significantly underutilized source of energy that holds immense potential for powering various micro-devices efficiently. Nonetheless, extracting high electric outputs from acoustic energy harvesters poses a considerable challenge. In this investigation, we adopt a novel one-step approach that combines electrospinning and electrospraying techniques to create a single-layer composite membrane. This membrane comprises a poly(vinylidene fluoride) (PVDF) nanofiber network with uniformly dispersed boron nitride nanosheet aggregates (BNNS-A), designed for sound energy harvesting purposes. Our device, measuring $3 \times 4 \text{ cm}^2$ with a PVDF/-BN configuration, yields peak voltage and current outputs of 174.2 V and 19.2 μA , respectively, when exposed to 115 dB, 230 Hz sound. Moreover, the incorporation of BNNS-A enhances the maximum peak power output by 30-fold compared to devices utilizing pure PVDF nanofibers. This study offers valuable insights into the endogenous triboelectricity mechanism in nanocomposites and introduces a promising approach for developing high-performance nanofiber acoustoelectric devices.

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The effect of electric fields on visible spin defects in hBN

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Some visible emitters in hBN have recently been shown to exhibit ODMR, but little is known about the atomic structure of these spin defects and the mechanism responsible for ODMR. In this study, we aim to obtain new insights by applying electric fields. Both vertical and planar electrode structures were fabricated which allow us to apply large electric fields to a hBN flake containing a high density of visible emitters. The effect of an electric field on the PL spectrum, the PL intensity, the excited state lifetime, the ODMR contrast, and the ODMR line shape, is studied. The results inform on-going efforts aimed at determining the nature of the visible spin defects.

Controlling and stabilizing the Charge State of Spin Defects in Hexagonal Boron Nitride

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Negatively charged boron vacancies (VB^-) in hexagonal boron nitride (hBN) have recently gained interest as spin defects for quantum information processing and quantum sensing by a layered material^[1]. However, the boron vacancy can exist in a number of charge states in the hBN lattice, but only the -1 state has spin-dependent photoluminescence and acts as a spin-photon interface. To establish control of the charge state, a custom confocal photoluminescent microscope attached to the chamber of a scanning electron microscope was constructed. This work demonstrates that under electron beam irradiation, the negatively charged VBs demonstrate deterministic and reversible charge state conversion ($\text{VB}^- \rightleftharpoons \text{VB}^0 + e^-$) with the excitation laser driving electrons to recombine with the defect, converting VB^0 back to VB^- . To control this behaviour, a layered hBN heterostructure was fabricated to control the rate of electron and hole injection into the defect area. The work displayed allows the stabilization of VB^0 defects into the useful VB^- charge state, enabling spin manipulation and optical readouts.

Fiber-integrated quantum sensors using color centers with optimal cavity interface

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Harnessing an optical fiber to quantum emitters provides considerable advantages given its convenience, compactness, and capability for distributed quantum information processing. There are substantial endeavors to interface solid-state emitters to an optical fiber, however, the coupling efficiency between the solid-state emitter and the optical fiber is limited by a restricted light extraction efficiency of host materials and small collection angles of fibers. Therefore, efficient interfacing solid-state emitters to the optical fiber remains a great challenge.

Recently, a hexagonal boron nitride (hBN) has received considerable interest by serving single photons and defect spins. Its unique two-dimensional configuration offers a transferable and integrable thin membrane. Moreover, their compatibility with well-established nanofabrication techniques has facilitated the integration of various nanophotonic structures. These benefits make the hBN an ideal quantum material to mate with optical fibers.

Here, we demonstrated a fiber-quantum sensor using an hBN membrane with an optimal optical interface, which consists of a fabricated hole-circular Bragg grating (hole-CBG) on the hBN flake integrated onto the optical fiber core (Fig. 1). From the results, we identified the cavity-enhanced spin-dependent photoluminescence via optical fibers. Our approach implements compact, practical quantum sensors for remote thermo-, magnetometry, essential for medical diagnosis and industrial inspections.

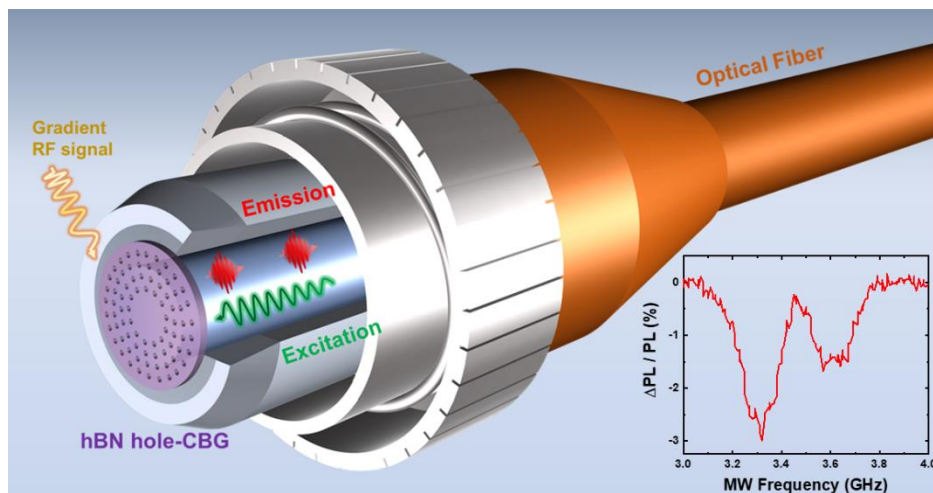


Figure 1 Schematic image of fiber-integrated hBN hole-CBG quantum sensor. (inset) ODMR spectrum of the V_B^- spin in hBN.

Quantum Sensing and Imaging of van der Waals Ferromagnet using Nitrogen-Vacancy Centers

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The nitrogen-vacancy (NV) centers in diamond are solid-state quantum emitters exhibiting unique spin and optical properties at room temperature. They are sensitive to magnetic fields, temperature, pressure, and other physical quantities, making them valuable as probes for sensing all of these physical quantities. In our work, we use NVs to form a magnetic microscope, with a high spatial resolution (~ 250 nm) limited by the diffraction limit and sensitivity of $<1 \mu\text{T}/\sqrt{\text{Hz}}$. This configuration is sometimes referred to as a Quantum Diamond Microscope (QDM) [1].

We use the QDM to reveal and understand the fundamental processes of magnetic domain pattern formation and their variation with temperature and external bias field, as well as characterizing the curie temperature (T_c) of recently discovered van der Waals (vdW) magnetic materials, namely Iron Germanium Telluride (Fe_5GeTe_2). We exfoliate these vdW materials down to a few nanometres. We observe that depending upon the thickness, their fundamental properties such as T_c , magnetization, domain pattern, etc. change.

We focus on measuring the T_c and imaging the domain structure of FGT flakes through out-of-plane magnetization. (Fig. 1). Our results [2] indicate structural features affecting magnetic orientation in these flakes, as well as a decrease in T_c as we are making the transition from bulk to 2D, which further decreases as the thickness of these 2D flakes decreases.

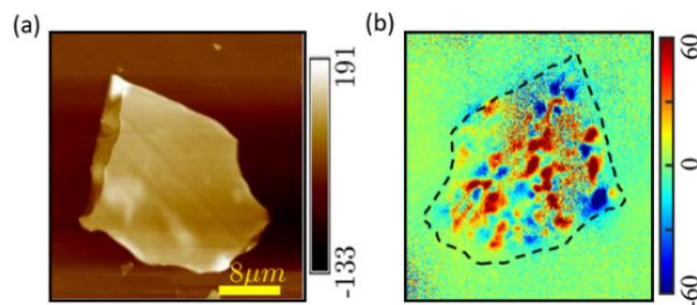


Figure 1. (a) Atomic Force Microscope (AFM) topography of the FGT flake. (b) Magnetic image of the same flake imaged by our QDM. Scale-bar units are nm (a) and $\mu\text{T}/\sqrt{\text{Hz}}$ (b).

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Exploring methods for creation of Boron-vacancies in hexagonal Boron Nitride exfoliated from bulk crystal

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Boron vacancies (VB^-) in hexagonal boron-nitride (hBN) have sparked great interest in recent years, due to their electronic spin properties. Since hBN can be readily integrated into devices where it interfaces a huge variety of other 2D materials, boron vacancies may serve as a precise sensor which can be deployed at very close proximity to many important materials systems. Boron vacancy defects may be produced by a number of existing methods, the use of which may depend on the final application. Any method should reproducibly generate defects with controlled density and desired pattern. To date, however, detailed studies of such methods are missing. In this paper we study various techniques, focused ion beam (FIB), electron irradiation and ion implantation, for the preparation of hBN flakes from bulk crystals, and relevant post-processing treatments to create VB^- s as a function of flake thickness and defect concentrations. We find that flake thickness plays an important role when optimising implantation parameters, while careful sample cleaning proved important to achieve best results.

Evaluation of hexagonal boron nitride in van der Waals junctions of 2D materials

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Hexagonal boron nitride (h-BN) crystals synthesized under high pressure and high temperature (HPHT) are known to have high quality and used in more than 200 research institutes as an atomically flat substrate for various 2D materials. However, all HPHT h-BN crystals have a distinct carbon(C)-rich domain at the core region, which contains a significant amount of C impurities (Figs. 1a,b). We found that this domain still exists after exfoliation and cannot be identified with an optical microscope or AFM (Figs. 1c,d), which suggests that the domain can be unknowingly incorporated into van der Waals (vdW) heterostructures. Considering the extensive use of HPHT h-BN, it is enormously important to investigate the C-rich domain in terms of its influence on adjacent 2D materials through vdW interface. To evaluate the influence of the C-rich domain on adjacent graphene, we first detected the domain region in h-BN flakes utilizing photoluminescence in the UV wavelength regime (Fig. 1d). Then, we fabricated graphene/h-BN heterostructures with graphene placed across the boundary of the domain, which enabled us to measure graphene on the domain and pristine regions simultaneously (Figs. 1e,f). Graphene on the C-rich domain showed lower carrier mobility (Fig. 1g) and higher carrier inhomogeneity n^* (Fig. 1h) than that on the pristine region and also exhibited bending in the Landau-fan diagram [1] (Fig. 1i). The plot of the energy at the bending point for each Landau level suggested the formation of an acceptor level at $E \sim 0.10$ eV above the Dirac point. To elucidate the role of C impurities in h-BN, we also conducted experiments using intentionally C-doped HPHT h-BN [2]. Finally, we present h-BN crystals synthesized at atmospheric pressure and high temperature (APHT) as an alternative to HPHT h-BN [3].

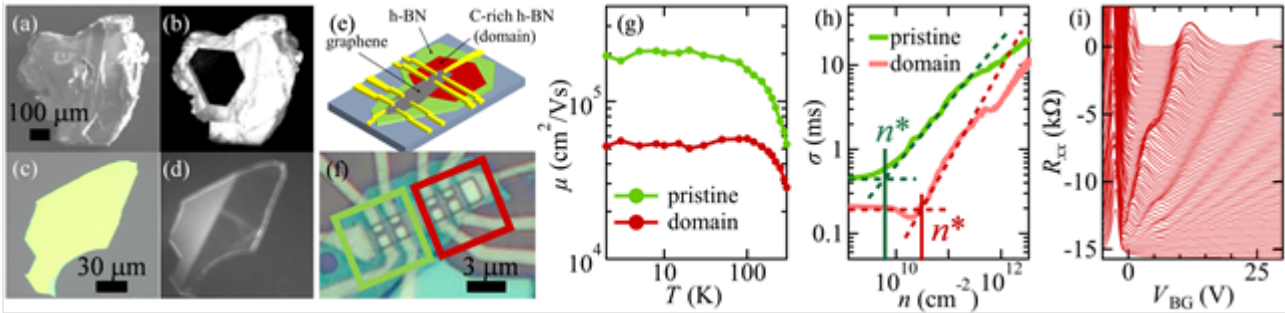


Fig. 1: (a) SEM and (b) CL (320 nm) images of an HPHT h-BN crystal. (c) Optical microscope and (d) PL (340 nm) images of an exfoliated HPHT h-BN flake. (e) Schematic and (f) photographic image of the device. (g) Temperature dependence of carrier mobility. (h) Extraction of carrier inhomogeneity n^* at $T = 2$ K. (i) R_{xx} vs. V_{BG} at $B = 0-9$ T obtained in graphene on the C-rich domain. Data are offset in proportional to B for clarity.

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Nitrogen isotope effects on hexagonal boron nitride

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Hexagonal boron nitride (hBN) is a wide indirect bandgap semiconductor which has gathered increasing attention due to its exceptional electronic and optical properties. Isotopic purification allows to improve some intrinsic properties of hBN, such as the phonon-polariton lifetime [1] and the thermal conductivity [2]. The effect of boron isotope purification on hBN was demonstrated in 2018 by Vuong et al [3].

Here, we extend the class of isotopically-purified hBN crystals to ¹⁵N, allowing us to have all the following configurations : h¹⁰B¹⁴N, h¹⁰B¹⁵N, h¹¹B¹⁴N and h¹¹B¹⁵N [4]. Firstly, we have studied the variations of the E_{2g}^{low} and the E_{2g}^{high} modes with the reduced mass thanks Raman spectroscopy, and then the impact of nitrogen isotope purification on the optical properties by UV-photoluminescence spectroscopy. The new h¹⁰B¹⁵N and h¹¹B¹⁵N configurations incorporate Raman and PL lines that are roughly the « mirror images » of h¹¹B¹⁴N and h¹⁰B¹⁴N with respect to h^{Nat}BN.

On the one hand, we have observed a progressive red-shift of the E_{2g}^{high} mode as a function of the reduced mass : boron and nitrogen atoms move in the opposite directions in the layer, similarly to a molecular vibrational mode, this result is coherent. On the other hand, for the interlayer shear mode, the E_{2g}^{low} mode, the behaviour is quite different and more difficult to interpret. However both results have been validated by DFT calculations. Finally, we have extracted the fundamental indirect exciton energy for each monoisotopic hBN crystal, that are consistent with the previous results [3].

Raman and photoluminescence spectroscopy reveal the achievement of ¹⁵N-purified hBN crystals of the same high-quality as for ¹⁴N, allowing to optimize the performance of devices based on hBN in the field of electronics or quantum technologies for example [5].

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Transport band gap measurement of large-area hBN by using direct and inverse photoemission spectroscopy

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We have investigated the transport and optical gap of hexagonal boron nitride (hBN) using the combination of UV photoelectron spectroscopy (UPS), inverse photoelectron spectroscopy (IPES), and reflection electron energy loss spectroscopy (REELS). Despite its importance in hBN, the experimental measurements of the transport band gap of hBN remains unclear, primarily due to challenges in synthesizing large-area sheets and its inherent insulating characteristics. To address this, we measured the transport gap of single to few layers of hBN synthesized on different large conducting substrates such as copper and ITO by using UPS and IPES. When combined with REELS measurements of optical gaps, we could infer concrete values of exciton binding energies for these samples. We found that the transport gap is roughly in agreement with calculated values reported previously, while the exciton binding energy is over 1 eV for single layer sample, which decreases as the layer thickness increases. We discuss the possible origins of these dependences.

Sensitivity Optimization of Boron Vacancy Centers in Hexagonal Boron Nitride

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The nitrogen vacancy (NV) center defect in diamond displays a photoluminescence (PL) dependent on the spin state, allowing for precision sensing of electromagnetic fields, temperature, and strain under ambient conditions. Despite all its success, the NV sensitivity is limited by the diamond host, which creates a separation between it and the sensing targets of at least a few nm to preserve good optical spin coherence properties. As an alternative, defects in wide band gap 2D materials have emerged as a promising platform. In particular, the recently discovered boron vacancy center defect in hexagonal boron nitride (hBN) shares several of the critical properties of the NV center: namely, a ground state electronic spin with long coherence time under ambient conditions; and optically detected magnetic resonance (ODMR). However, many of the sensitivity limitations of boron vacancies remain largely unexplored. In this poster, we discuss characterizing the sensitivity of the boron vacancies, as well as the effect of hBN flake size. Additionally, we provide an overview of lock-in and other measurement techniques to help improve the overall sensitivity of the boron vacancy center to electromagnetic fields by suppressing added noise. We also have ongoing investigations to optimize sensitivity: e.g., using optical excitation at 475 nm wavelength rather than 532 nm to increase boron vacancy PL.

Toward Nanoscale NMR Spectroscopy using the Boron Vacancy Quantum Defect in hBN

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(Dated: February 8, 2024)

The use of optically active quantum defects in solids as magnetic field sensors is a leading technique for nuclear magnetic resonance (NMR) spectroscopy and imaging of nanoscale samples under ambient conditions. For example, the Nitrogen-Vacancy (NV) center in diamond has been used for NMR detection of single proteins. However, the spin and optical properties of NV centers degrade when closer than about 10 nm from the diamond surface, limiting sensitivity as well as spectral and spatial resolution. Here we describe efforts to develop an alternative nanoscale NMR sensor using the negatively charged boron vacancy (V_B^-) in hexagonal Boron Nitride (hBN). Theoretical predictions and measurements to date indicate that the spin and optical properties of the boron vacancy do not significantly degrade in few layer hBN, potentially allowing a standoff distance < 1 nm between the boron vacancy and sensing targets on the hBN surface. We describe ongoing work to address key technical challenges, e.g., sample confinement to ameliorate broadening of the measured NMR linewidth due to nanoscale diffusion effects; sample localization with individual boron vacancy sensors; and experimental techniques to hyperpolarize the sample nuclear spins and/or perform high-resolution NMR spectroscopy with statistically polarized spins. We propose a complete nanoscale NMR protocol using single V_B^- in hBN, including relevant pulse sequences, sensitivity calculations, and sample confinement strategies. We account for unconventional diffusion dynamics in the flow-restricted nanoscale regime and calculate its effects on the expected NMR signal. Lastly, we identify potential sample targets for nanoscale NMR applications best suited to this novel approach.

Spatially-resolved UV-C emission in epitaxial monolayer boron nitride

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The electronic bandgap of hexagonal boron nitride (hBN) is calculated to be indirect in its bulk form and direct at the monolayer limit. Experimental evidence for indirect-gap bulk hBN was brought by optical spectroscopy in the UV-C spectral range [1,2], and independently confirmed by electron energy loss spectroscopy [3]. The indirect-direct crossover of the bandgap was first experimentally demonstrated by reflectivity and photo-luminescence (PL) spectroscopy in large-scale monolayer hBN epitaxially grown on graphite by high-temperature molecular beam epitaxy (HT-MBE) [4], and then reproduced by hyperspectral imaging in exfoliated hBN flakes at low temperature [5], by means of a scanning confocal cryomicroscope operating at the diffraction limit at wavelengths around 200 nm. A specific issue of PL experiments in monolayer hBN under quasi-resonant excitation is the coexistence of resonant Raman scattering and PL which in turn complicates the interpretation of optical spectroscopy when characterizing atomically-thin hBN in the UV-C range.

Here we report hyperspectral imaging in the UV-C spectral domain in epitaxial monolayer hBN, under quasi-resonant laser excitation. Thanks to our spatially-resolved measurements of the UV-C emission, we spatially isolate the distinctive contributions of resonant Raman scattering and PL. We provide a quantitative interpretation of our experiments by means of a weighted average of resonant Raman scattering and PL, from which we extract the maps of their respective amplitudes in our epitaxial monolayer hBN sample. No spatial correlation of the resonant Raman scattering and PL amplitudes is observed. The PL signal consists in a singlet at ~ 6.045 eV, with spatial variations of the PL energy around ~ 10 meV, thus revealing that the inhomogeneous broadening is lower than the average PL linewidth of ~ 25 meV, a value close to the radiative limit determined by reflectivity experiments. Our methodology provides an accurate framework for assessing the opto-electronic properties of hBN in the prospect of scalable hBN-based devices fabricated by epitaxy.

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Remote moire effect engineering using the twisted hBN

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By stacking two-dimensional materials with marginally twisted angles, it allows the moire potential at the interface. Recently, many studies have been conducted in which the moire potential affects other remote two-dimensional materials. Hexagonal boron nitride (hBN) has atomically flat surfaces and it allows the intrinsic properties for the other materials. When two hBNs are aligned parallel with slightly twisted angles, it forms the periodic AB-BA domains with different potential. Here, we observed the monolayer MoSe₂ photoluminescence properties on the twisted hBN. We measured around 100 meV moire potential modulation in monolayer MoSe₂ formed by twisted hBN using Kelvin probe microscope (KPFM). And we probe the moire effect by placing twisted hBN with various twist angles.

High electric field vertical tunneling transports in hexagonal boron nitride

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Hexagonal boron nitride (hBN) is a van der Waals material with a hexagonal structure like graphene, but hBN is a wide bandgap material with a bandgap exceeding 6 eV. Due to the high thermal stability and atomically flat surface of single crystal hBN, hBN has emerged as an ideal 2D dielectric material for the encapsulation layers to protect other 2D materials and the excellent tunneling barrier materials for tunneling devices. Here, we realized the vertical tunneling device with hBN for tunneling barrier and graphene electrodes using van der Waals pick-up methods to get atomically clean interfaces. We observed the ideal tunneling behavior transition between direct tunneling and Fowler-Nordheim (FN) tunneling from hBN heterostructures under an applied electric field above 0.7 V/nm. Under a high electric field condition to hBN tunneling device, electric field-induced defects such as vacancies and impurities incorporated in hBN significantly change the tunneling behaviors. We also probed the carbon color center-mediated vertical tunneling transport and significantly enhanced the breakdown field in the hBN tunneling architectures.

Hexagonal boron nitride surface engineering for remote modulation doping

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Hexagonal boron nitride (hBN) is a two-dimensional (2D) material known for its excellent thermal conductivity, electrical insulation properties, and chemical stability. Due to its unique properties, hexagonal boron nitride is widely utilized in research and as an excellent encapsulation material for 2D materials studies. After developing the van der Waals pick-up method with hBN and edge-contact method in graphene devices exhibited the theoretical phonon-scattering limit at room-temperature and ballistic transport up to 15 micrometers at low temperature^{[1], [2]}. Previously, surface engineering has been investigated to actively control defects in hBN, aiming to modulate its optical and electrical properties. Moreover, studies have investigated the introduction of deep-level states into the band gap of hBN through oxygen plasma treatment^[3]. In this study, we present the engineering of the n-type remote modulation doping effect in hBN/Gr/hBN devices by controlling the O₂ plasma-induced surface engineering of the top hBN layer and the trap states in hBN. We observed the degree of doping effect by differing O₂ plasma treatment times in graphene heterostructures and achieving the Ohmic edge contact with low contact resistance (~ 150 Ohm.μm) at low temperature, resulting in the observation of quantum transport such as the quantum Hall effect and Hofstadter butterfly. These results pave the way for remote modulation doping strategies and contact engineering to develop high-performance 2D optoelectronic devices.

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Defect engineering in CVD-Grown Hexagonal Boron Nitride for Quantum Photonic Applications

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Hexagonal boron nitride (hBN) has recently emerged as an ideal platform for quantum photonic applications, because they can host isolated defects as a source of single-photon emitters (SPEs) and also optically addressable spin defects as quantum sensors. Large-area hBN films grown by chemical vapor deposition (CVD) are appealing due to their scalability of the above-mentioned applications. However, CVD-grown hBN are usually defective with strong defect emission backgrounds. Here we report using high-quality CVD grown multilayer hBN as the host material to create SPEs and boron vacancy (V_B^-) ensembles. The multilayer hBN were grown by atmospheric-pressure CVD on molten Cu, resulting in a 10-nm-thick large-area hBN film with a low surface roughness of ~ 0.64 nm. Defects for SPEs were created by thermal annealing and UV ozone treatments. The defect emissions show a strong zero phonon line (ZPL) near 575 nm with a typical linewidth < 10 nm (lowest ~ 3 nm). Photon correlation measurements for most defects show $g^{(2)}(0) < 0.5$, indicative of SPEs. On the other hand, we created spin-active defects by ion implantations. The induced defects show a broad emission band centered at 850 nm, indicative of V_B^- ensembles. We have performed optically detected magnetic resonance (ODMR) measurements to study the electronic structure of V_B^- . Magnetic-field dependent ODMR measurements show two dips near 73 mT and 130 mT, corresponding to the excited-state and ground-state level anti-crossing, respectively. We have also measured high-field ODMR and their angle dependence to characterize the axial symmetry and g-factor anisotropy of the defects. Our results demonstrate that CVD-grown large-area multilayer hBN films can be an ideal host material for SPEs and spin-active defects.

High-efficiency deep ultraviolet emitting from hexagonal boron nitride heterostructure

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Two-dimensional materials are recognized for their extensive light emission capabilities; however, their ability to emit ultraviolet (UV) light remains relatively unexplored. This study examines hexagonal boron nitride (hBN), a material with a ultra-wide bandgap of 6.2 eV, demonstrates potential for emitting a diverse range of UV wavelengths. In our research, we have developed a light-emitting device that utilizes a van der Waals heterostructure with graphene electrodes and hBN tunneling layers. We also observed the carbon color centers are created by strong electric field and their corresponding emissions. Carbon color centers in hBN enhance charge carrier injections to tunneling process, resulting in enhance the light emission efficiency.

UV defect emitters in thin hBN

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Room temperature defect quantum emitters in hexagonal boron nitride (hBN) have emerged as a source of considerable scientific interest. Recent studies have demonstrated the ability to fabricate reliable single-photon sources in thin film hBN with reproducible emission properties in all spectral regions [1], suitable for applications in quantum technologies. While such quantum emitters have been extensively studied in the visible and near-IR, this work focuses on exploring the UV luminescence of hBN point defects at 4.1 eV, likely caused by carbon substitutions at nitrogen sites [2-3].

We present a comprehensive study of emission intensity, line width, and carrier dynamics as a function of temperature for UV emitters in both carbon-doped and as-grown exfoliated and MBE-grown thin films. A frequency-quadrupled titanium-sapphire laser at 200 nm is used for pulsed above-bandgap excitation and compared to sub-bandgap pulsed and continuous wave excitation. A direct dependence between the luminescence of the defect and its surroundings is explored to establish a link between the properties of the 2D material samples and the electron-phonon coupling. These results are corroborated by the correlation of micro-Raman and micro-PL mapping. Furthermore, the photon statistics of these centres are studied by μ -PL autocorrelation spectroscopy in search of UV single photon emitters. In addition, we discuss the recently proposed correlations between UV and blue emitters using photoluminescence excitation spectroscopy.

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Hexagonal Boron Nitride-Based Quantum Key Distribution with Room Temperature Single Photon Emission

Helen Zhi Jie Zeng,^{1*} Ali Al-Juboori,¹ Minh Anh Phan Nguyen,¹ Xiaoyu Ai,² Adam Bennet,³ Arne Laucht,² Alexander Solntsev,¹ Milos Toth,^{1,4} Richard P. Mildren,³ Robert Malaney,² and Igor Aharonovich^{1,4}

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Solid state materials are the fundamental cornerstone for quantum-based technologies, such as quantum key distribution (QKD)[1]. Hexagonal boron nitride (hBN) is capable of hosting atomic defects which can emit high-purity single-photons at room temperature [2]. Single photon sources (SPS) is integrated in hBN with solid immersion lenses (SILs) to increase the signal-to-noise ratio and combat the problem of collection efficiency and demonstrate a discrete-variable quantum key distribution system.

SIL made of 1mm diameter cubic zirconia hemispheres were used. Solvent-exfoliated hBN flakes were deposited onto the flat side of the SIL and emitters were activated using high-temperature annealing. Optical characterisation was performed on a custom-built portable confocal microscope (Fig. 1a) with 532 nm and 515 nm excitation wavelengths for continuous wave and pulsed measurements respectively.

A characteristic SPS shown with $g(2)(0)$ value as low as 0.07 (Figure 1a inset & b). Single-photon rates of 107 Hz was achieved while maintaining photostability over extended durations and adaptability to a portable, lightweight setup (Figure 1a & c). A finite-key BB84 QKD system operating with hBN defects (Figure 2a & b), producing secret keys of approximately 70,000 bits at a QBER of 6% and ϵ -security of 10^{-10} . To demonstrate QKD utility we securely transfer an image from Alice to Bob (Figure 2c-e).

Our source performs with high brightness exceeding 4×10^5 photon/s under pulsed excitation and operates under ambient conditions. This work puts forward a robust and scalable implementation of QKD using triggered room temperature SPSs based on solid materials.

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Figures

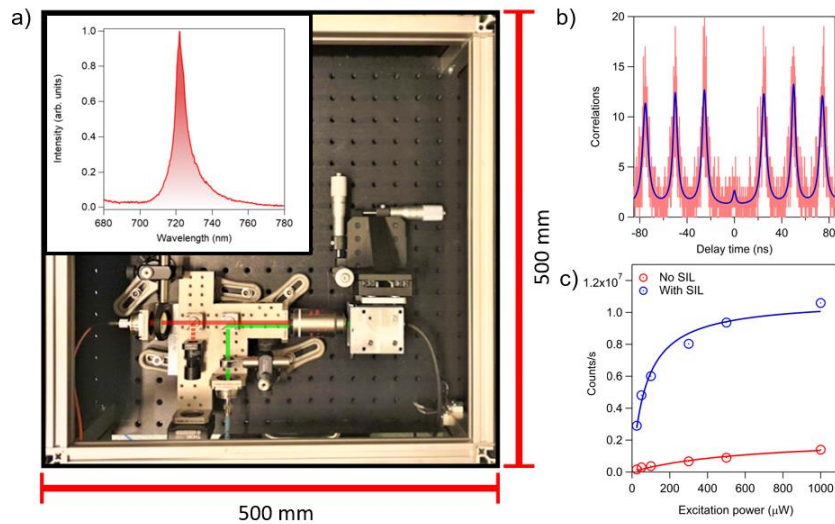


Figure 1. a) Top-down photograph of the confocal setup with inset characteristic SPS PL spectra. b) Pulsed second-order autocorrelation measurement showing a $g(2)(0)$ value of 0.07. c) Power-dependence of emitter with and without SIL.

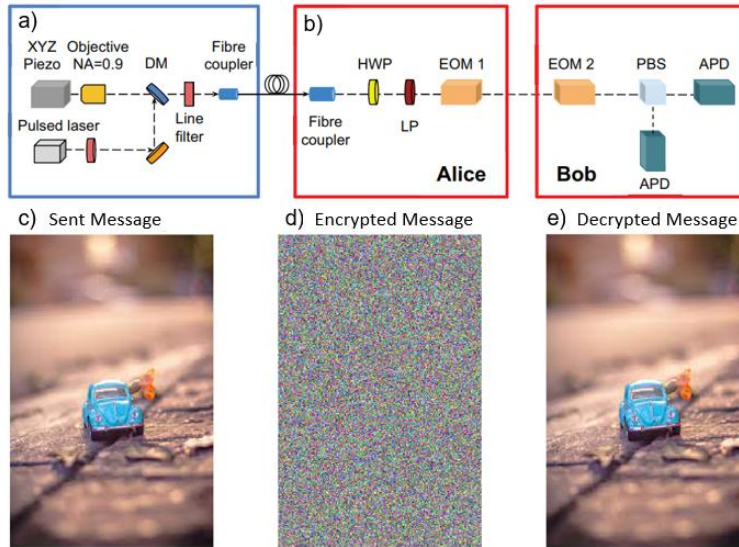


Figure 2. a) Schematic diagram of the single-photon source. DM, dichroic mirror. b) Schematic diagram of the QKD setup. EOM, electro-optic modulator; LP, linear polariser; APD, avalanche photodiode; PBS, polarizing beam splitter; HWP, half-wave plate. c) Original image, d) image encrypted with Alice's secure key, and e) the decrypted image after decoding it using Bob's secure key.

Magnetic Field Sensitivity Optimization of Negatively Charged Boron Vacancy Defects in hBN.

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Optically active spin defects in hexagonal boron nitride (hBN) have recently emerged as compelling quantum sensors hosted in a two dimensional (2D) material. The photodynamics and sensitivity of spin defects are governed by their level structure and associated transition rates. The photodynamics of the negatively charged boron vacancy (V_B^-), while being the most widely-studied spin defect in hBN, is yet to be fully understood. The V_B^- defect can be simplified into a 5-level system (Figure 1 (a)) with intersystem crossing rates defining the internal mechanisms that allow high resolution and high sensitivity quantum sensing.

Optical and microwave pump-probe measurements were used to characterize the relaxation dynamics within the energy levels of the V_B^- defect. A 5-level model was used to deduce transition rates that give rise to spin-dependent V_B^- photoluminescence, and the lifetime of the V_B^- intersystem crossing metastable state. The obtained rates are then used to simulate the magnetic field sensitivity of V_B^- defects (Figure 1 (b)) and demonstrate high resolution imaging of the magnetic field generated by a single magnetic particle using optimal sensing parameters predicted by the model. The results reveal the rates that underpin V_B^- photodynamics, which is important for both a fundamental understanding of the V_B^- as a spin-photon interface and for achieving optimal sensitivity in quantum sensing applications.

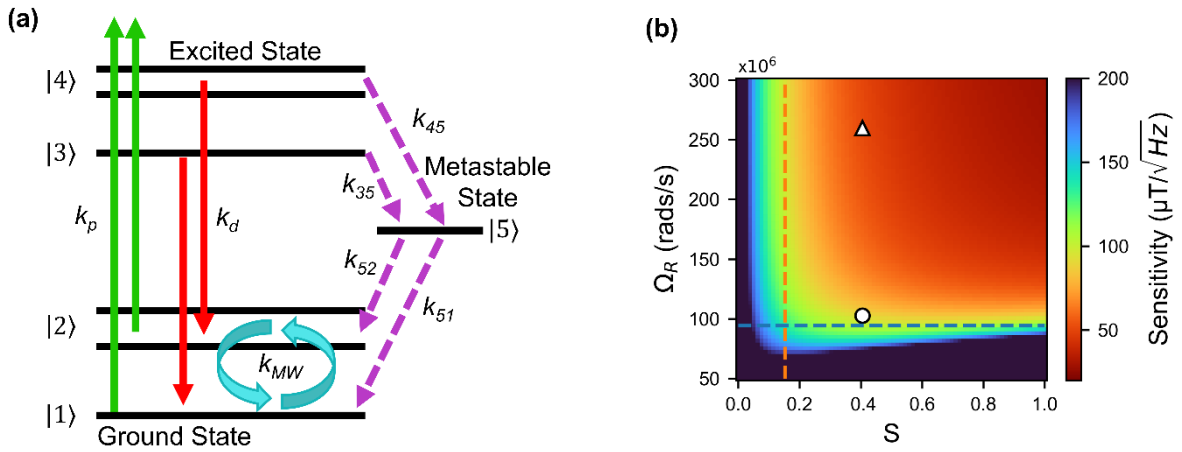


Figure 1. (a) Simplified 5-level energy diagram of the VB- defect. The $m_s = +1$ and $m_s = -1$ spin states are combined into $|2\rangle$ and $|4\rangle$ for the ground and excited state, respectively. Arrows indicate various transitions between sublevels. **(b)** A map of simulated sensitivity versus both relative laser power (S) and Rabi frequency (Ω_R). Constructed with rates acquired from the 5-level model.

Plasmonic lattices-assisted emission enhancement and optimized creation of blue colour centre in hBN

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The B-centre defect in hexagonal boron nitride (hBN) that emits at 436 nm shows promise as a single photon source for photonic quantum technologies. Activating defects with focused electron irradiation in scanning electron microscopes (SEM) offers solutions to the uncontrolled fabrication method required for emitters in hBN. For some applications including on chip photonic resonators or plasmonic cavities, hBN flake thickness also plays a key role. In this work, we demonstrate a method to incorporate the light sources into plasmonic lattices, and robust procedures based on plasma treatments to increase the activation efficiency of B-centres in thin (<10 nm) hBN. The utilization of aluminium metal is essential due to the blue wavelength. As a result, trench resonator geometry was employed to systematically move the hBN and emitter to the plasmonic hotspot. We achieved moderate enhancement in photoluminescence emission by six times from blue emitters in ~ 50 nm thick flake assisted by plasmonic-coupling (shown in Figure 1).

We present the limitation of engineering B-centre in thin flake (< 20 nm) due to the contaminants such as water and hydrocarbons as residual polymer residues after exfoliation and flake transfer procedures. We achieved the activation of emitters in hBN flakes as thin as 8 nm with the optimized cleaning procedure before irradiation using plasma cleaning in the SEM chamber (shown in Figure 2(a)). We also report on robust procedures based on UV/Ozone treatment after the irradiation to improve signal-to-background noise ratio of the B centres (shown in Figure 2(b-f)). The optical characterisation of the emitter is also performed. Our methodology is key to enable scalable engineering of B-Centres for on chip quantum photonic applications.

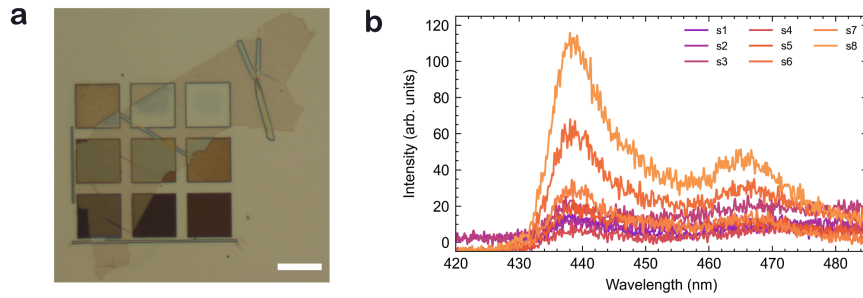


Figure 1. (a) Optical image of 50 nm thick hBN flake integrated on aluminium plasmonic lattices. (b) PL spectra of the B-Centres located on top of plasmonic lattices. Enhancement is observed for spots S7 and S8, likely due to the overlap of the formed emitters and the nanotrench.

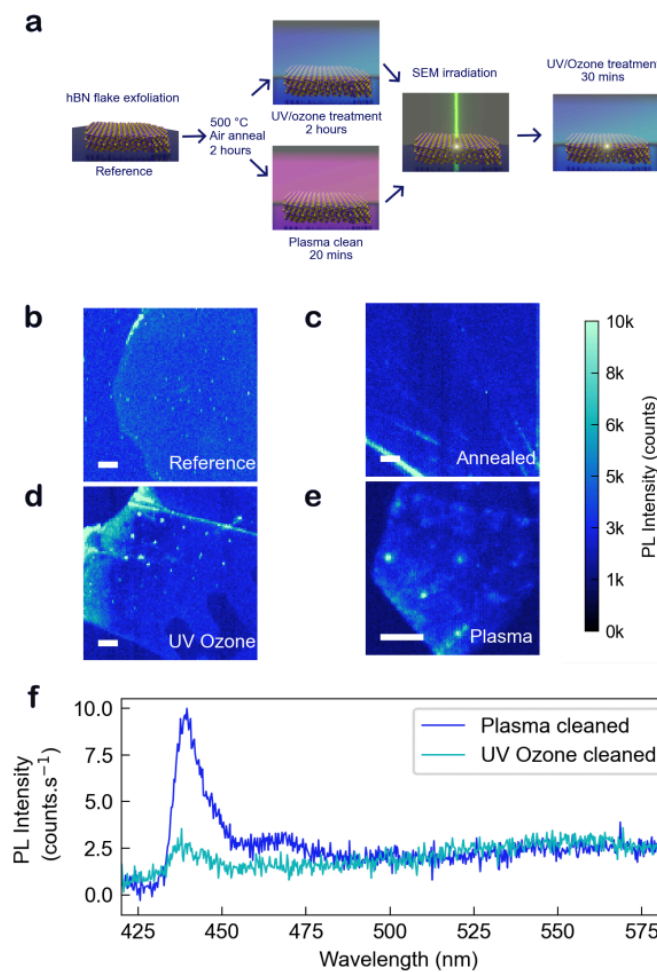


Figure 2. Comparison of pre-cleaning methods used before activation of B-centres in hBN flakes with thicknesses < 20 nm. (a) Schematic of the cleaning procedures and the activation of B-centres by a focused electron beam. (b-e) Confocal PL maps of hBN flakes comparing the pre-cleaning treatments; reference, air annealed at 500 °C, UV/ozone treated and in-situ plasma cleaned. The scale bars in all maps are 5 μm . (f) PL spectra from emitters in UV/ozone and plasma treatments.

Resonant Spectroscopy of B-Center Quantum Emitters in hBN

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Abstract: A site-specific fabrication technique was used to produce arrays of blue quantum emitters in hBN for a cryogenic spectroscopy study. We characterise temperature dependent linewidth, identify processes that limit emitter coherence, and observe Rabi oscillations.

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Solid-state two level systems have the potential to be efficiently integrated into scalable quantum technology devices, enabling opportunities in enhanced sensing, secure communication, and quantum computation [1–3]. Interest is growing in the use of layered two dimensional materials such as hexagonal boron nitride (hBN), given their atomically thin nature. The exploration of defects in hBN has shown this platform is capable of hosting both spin [4–6] and single photon emitters (SPEs) [7–10], which could provide the basis for a spin-photon interface for use in quantum information distribution [11].

Despite progress, achieving desirable photonic capabilities from an SPE in hBN continues to impose significant experimental challenges. Most notably, hBN hosts numerous SPEs with a broad range of emission wavelengths, and most defect fabrication methods are ineffective at generating SPEs consistently with a small variance in emission wavelength. An exception to this problem is a recently-developed electron beam irradiation technique for the generation of so-called blue emitters with an emission wavelength at ~ 436 nm [12–14]. The technique is site-specific, it works reliably with high quality hBN crystals and the emitters can be produced en masse – all of which make the blue emitters appealing for studies of SPE spectral behavior and coherence properties.

In this work, we characterize the non-resonant photoluminescence properties of the blue emitter at cryogenic temperatures. Over two dozen sites, we observe an ultra-narrow distribution of the zero phonon line at ~ 436 nm, together with strong linearly polarized emission. We employ resonant excitation to characterize the emission lineshape and find spectral diffusion and phonon broadening contribute to linewidths in the range 1-2 GHz. The emitter linewidth is found to have a cubic dependence on temperature, and broadening mechanisms are prevalent even at 5 K. An excited state lifetime of 2.27 ns is measured using a pulsed off-resonant laser, providing an estimate of 70 MHz for the natural emission linewidth.

Most significantly, Rabi oscillations are observed at a range of resonant excitation powers (see Fig. 1). A second order correlation model allows the power dependent pure dephasing rate to be extracted, and this provides a means to calculate the coherence time of the emitter. Under 1 μ W resonant excitation a coherent superposition is maintained up to 0.90 ns. This result is a key requisite capability for these blue emitters to be considered as a viable platform for the development of scalable quantum technologies in hBN. In particular, the ratio of the coherence time to the lifetime is important for experiments involving two photon interference for entanglement distribution.

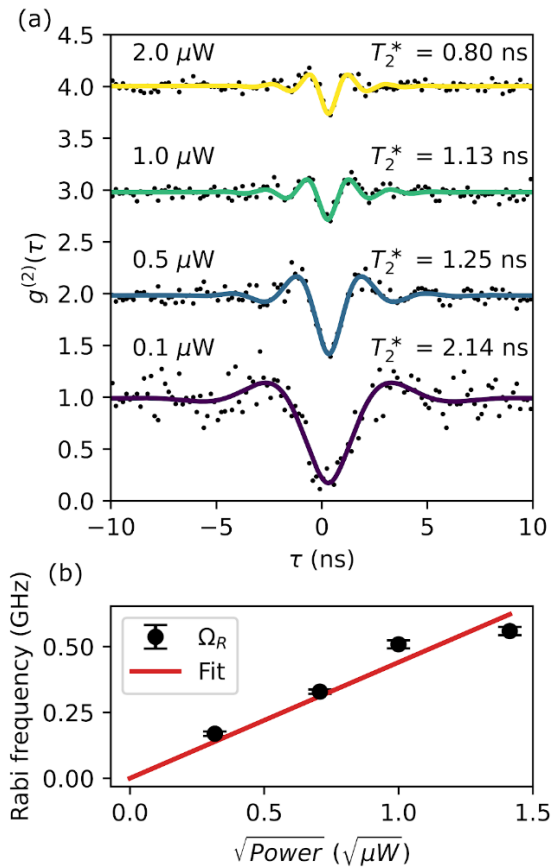


Fig. 1. Coherence properties. a) Second order correlation measurement collected over a range of excitation powers, and the fitted function demonstrating the presence of Rabi oscillations. Each plot is vertically offset for clarity. Values of the pure dephasing time T_2^* are extracted from the fit. b) The Rabi frequency extracted from the fit in (a) is plotted against the square root of the excitation power, and a linear function is used to fit the data.

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Towards Boron Nitride Nanotube Optical Emitters in Sensing Applications
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Since their introduction in the 90s, boron nitride nanotubes (BNNTs) have attracted interest for their chemical and structural properties that are comparable or superior to carbon nanotubes. Their low density and high surface to volume ratio points towards a multitude of sensing and filtering applications, as well as their high Young's modulus and tensile strength for reinforcing filler applications. Optically active defects in BNNTs take advantage of these unique structural properties in applications that previous lattice arrangements would not allow them to be used. Hexagonal boron nitride (hBN) is host to a spin- and optically-active defect with an emission centred around 800 nm, namely the negatively charged boron vacancy defect (V_B^-). This defect is created deterministically and uniformly in ensembles, hosting a broad emission that can be coupled to photonic structures.

In this work, the (V_B^-) defect was created in BNNTs via N^+ ion irradiation, and its generation parameters were optimised. V_B^- exists in flat, single layers of hBN, so its novel structure in BNNTs was investigated under TEM, along with photoluminescence and ODMR optical characterisations.

BNNTs can be placed on an atomic force microscope (AFM) tip for magnetic field sensing applications. Previous works have demonstrated BNNTs on an AFM tip with spin-active ~ 600 nm emitters [1], but this work looks to fabricate this with the more reliably created V_B^- defect.

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Photodynamics of electrically driven isolated colour centres in van der Waals semiconductors

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Single photon sources also referred to as quantum emitters in two-dimensional (2D) materials, such as hexagonal boron nitride (hBN) offer great prospects for integration in photonics, sensing and quantum communications devices. hBN hosts a large range of quantum emitters in the visible wavelength range that display good qualities such as purity, stability [1] and optically active spin [2], which can be employed in sensing and storage applications. Demonstration of electroluminescence (EL) of these lattice defect colour centres is an important milestone on the path to scalable on-chip quantum technologies. Although their quantum nature was not yet realised, this work discovers novel key achievements in the field, such as new method of activating isolated colour centres in hBN, first electrically driven excitation that doesn't utilise a p-n junction device, and mechanisms behind colour centres operation at cryogenic and room temperatures.

By exploiting the van der Waals nature of 2D materials we are able to design heterostructures that build on quantum properties of charge carriers, such as tunnelling. Through a series of electrical measurements we derive that the driving mechanism behind colour centres excitation follows Fowler-Nordheim tunnelling model which injects charges into band edges of hBN. This carrier injection leads to simultaneous excitation of multiple colour centres that emit light across a wide range of frequencies, spanning from ultraviolet to near infrared. We report prolonged operation of EL devices at cryogenic and room temperatures and examine optical characteristics of isolated colour centres, which display minimal fluctuations over time in parameters such as emission centre wavelength, full width at half maximum and intensity. Polarization measurements showed single optical transitions for many of observed emissions and spectral characteristics analysis directly related them to common hBN quantum emitters. Additional data in this presentation talks about creation dynamics of these colour centres and their behaviour under applied electric field during optical excitation.

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Correlating Defect Emission with Infrared Near-Field Imaging in Strained Hexagonal Boron Nitride

Ryan Kowalski
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Point defects in hexagonal boron nitride (hBN) represent promising candidates for quantum information science technologies. The substantial electronic bandgap (~ 6 eV) of hBN offers protection for electron defect states against external disturbances like thermal noise, enabling robust, room-temperature single photon emission in the visible spectrum within a two-dimensional structure. Nevertheless, strain in the surrounding crystal lattice impacts the emission characteristics of hBN single-photon emitters (SPEs), causing reversible photoluminescence (PL) wavelength shifts,¹ which may serve as a tuning mechanism. Strain has also been linked to near-deterministic SPE generation,² a desirable trait for on-chip device fabrication.

In this study, we transfer large-area hBN flakes onto silicon nanopillar arrays to induce bright emission and investigate local strain gradients at the nanopillar sites. We correlate the PL emission with infrared scattering-type scanning near-field optical microscope (s-SNOM) images, a nanoscale spectroscopic tool for probing local dielectric variations at extremely sub-diffractive length scales (< 20 nm). PL intensity mapping of transferred hBN (Figure 1a) reveals preferential emission at nanopillar sites, especially around edges where strain profiles peak. An s-SNOM image of an individual nanopillar (Figure 1b) illustrates hBN conforming around it. Nanoscale Fourier transform infrared (nano-FTIR) spectra are measured across the top of the nanopillar, where each point corresponds to a fully interferometric infrared spectrum displayed in Figures 1c and 1d. The distinct peak at 1360 cm^{-1} corresponds to the transverse optic (TO) phonon mode, which spectrally shifts at the positions along nanopillar edges. This is consistent with previous reports of strain in hBN nanobubbles.³ This research aims to identify the relationship between hBN SPEs and its local lattice through nanoscale vibrational spectroscopy, an area warranting further investigation.

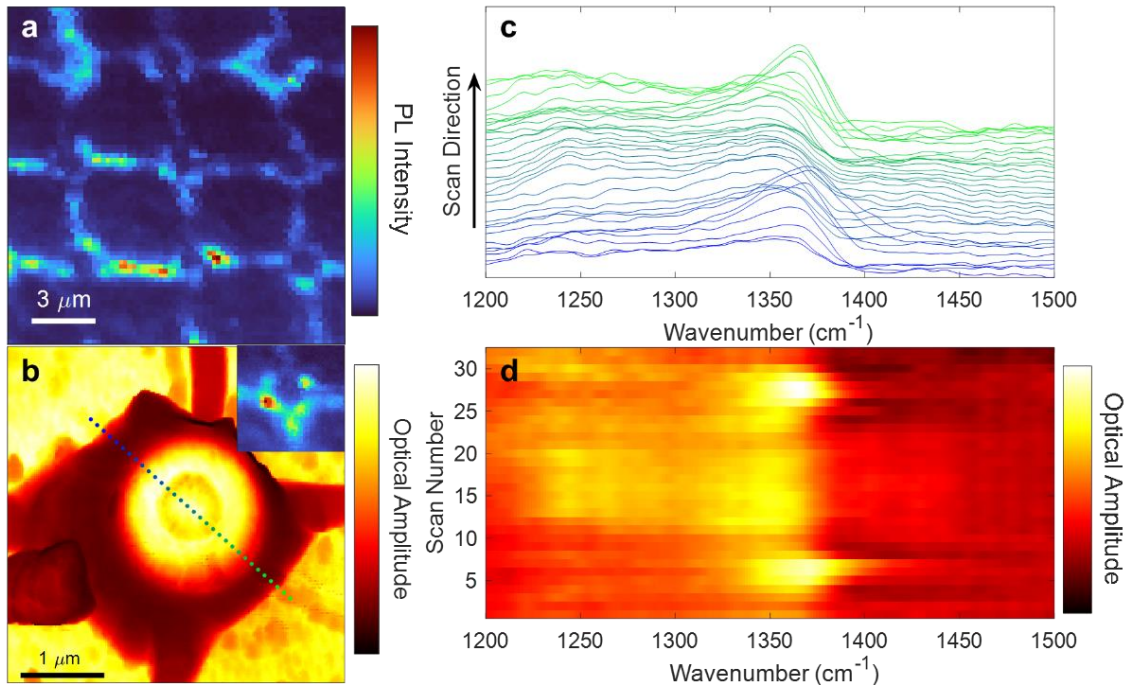


Figure 1: a) PL intensity map of hBN transferred on a Si nanopillar array ($d=1500$ nm, $h=350$ nm). Images are taken from a lab-built scanning optical microscope (532 nm, 0.7 NA) with a ~ 1 μm laser focus at 100 μW . b) Integrated ($1200\text{-}1500$ cm^{-1}) infrared s-SNOM image of a single nanopillar. Colored dots indicate the positions of individual interferometric nano-FTIR spectra taken across the pillar, the scan direction begins in the top left (blue) and ends in the bottom right (green). Inset: PL map of nanopillar. c) Nano-FTIR amplitude spectra corresponding to colored dots in (b). d) Contour plot of amplitude across (b), scan 0 (32) correspond to the blue (green) dots.

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Coherent control of an ultrabright single spin in hexagonal boron nitride at room temperature

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Abstract: Hexagonal boron nitride (hBN) is a remarkable two-dimensional (2D) material that hosts solid-state spins and has great potential to be used in quantum information applications, including quantum networks. However, in this application, both the optical and spin properties are crucial for single spins but have not yet been discovered simultaneously for hBN spins. Here, we realize an efficient method for arraying and isolating the single defects of hBN and use this method to discover a new spin defect with a high probability of 85%. This single defect exhibits outstanding optical properties and an optically controllable spin, as indicated by the observed significant Rabi oscillation and Hahn echo experiments at room temperature. First principles calculations indicate that complexes of carbon and oxygen dopants may be the origin of the single spin defects. This provides a possibility for further addressing spins that can be optically controlled.

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Nanoporous atomically thin ceramic membranes for energy and healthcare

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Atomically thin membranes present potential for disruptive advances in separations for energy and healthcare applications. Here, I will discuss our recent advances in growth of atomically thin h-BN membranes and fabrication of fully functional membranes. Specifically, I will focus on quantifying sub-nanometer scale defects in monolayer h-BN membranes over centimeter scale areas that have remain inaccessible via conventional characterization techniques. Next, I will discuss the fabrication and evaluation of atomically thin membranes for energy application and healthcare applications. We demonstrate fully functional atomically thin ceramic membranes that outperform state-of-the-art conventional membranes by order of magnitude while maintaining adequate separation quality or doing better. Finally, I will discuss the transition of these technologies from the lab to the commercial arena.

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Growth of wafer-scale, high-quality, and uniform multilayer hBN film on liquid Fe₂B for high-performance of 2D heterostructure

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Multilayer hexagonal boron nitride (hBN) serves as an ideal substrate for realizing the intrinsic properties of two-dimensional (2D) materials in diverse electronic devices [1]. There is a growing demand for wafer-scale, high-quality, and uniformly grown multilayer hBN films for various scalable applications. In this study, we present a facile method for achieving high-quality and wafer-scale uniform growth of multilayer hBN films on liquid Fe₂B via chemical vapor deposition. The resulting multilayer hBN film demonstrates remarkable uniformity, featuring a large grain size of approximately $167.81 \pm 10.98 \mu\text{m}$, with a full width at half maximum value of around 9.8 cm^{-1} for the E_{2g} peak. These values are comparable to those of exfoliated hBN [2]. Furthermore, the grown hBN films are employed in single-crystal graphene and MoS₂ films to investigate scalable applications. The optical and electrical properties based on the 2D heterostructures showcase intrinsic and high performance, demonstrating the high-quality of the grown multilayer hBN film at the wafer scale.

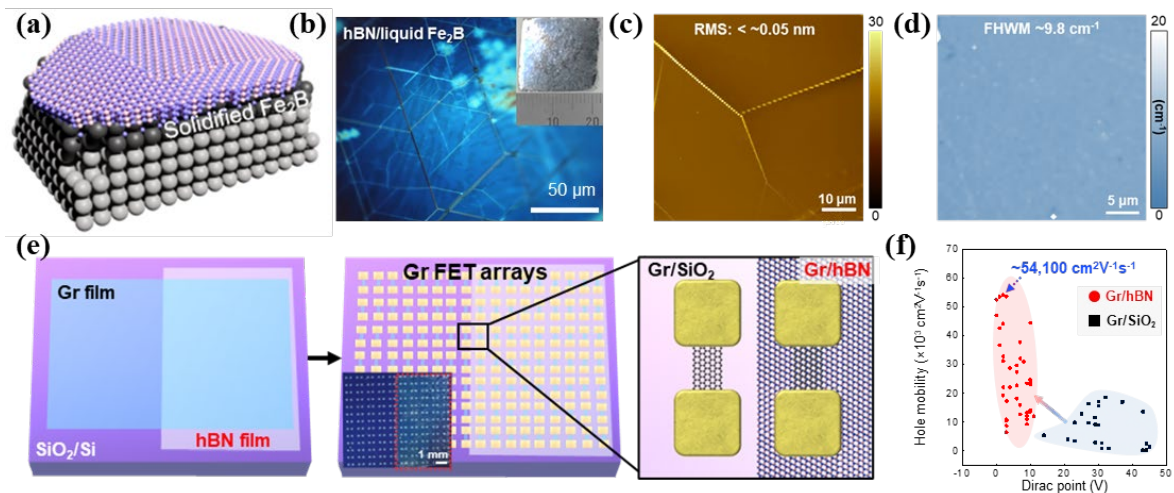


Figure. (a) Scheme for growth of multilayer hBN on liquid Fe₂B, (b) optical micrograph of the as grown multilayer hBN film, (c,d) AFM and Raman FWHM mapping (E_{2g}) images of the transferred multilayer hBN film, (e) scheme for the preparations of graphene (Gr) field effect transistor arrays, (f) distributed plots of the carrier mobilities of Gr FETs.

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