Abstract booklet

Invited

Controlled CVD growth of multilayer hBN for 2.5D applications

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We have been studying the controlled CVD growth of hexagonal boron nitride. By developing our original epitaxial CVD growth of monolayer graphene on Cu(111)/sapphire [1-3], we investigated the hBN growth on the Cu(111)/sapphire substrates. The Cu(111) allowed us to synthesize epitaxially aligned triangular grains of hBN [4]. We can also make sub-millimeter triangular hBN grains on Ni(111)/sapphire by increasing the CVD temperature [5]. However, as the hBN grown on Cu(111) and Ni(111) is monolayer, it is not thick enough for the application to 2D insulators.

Recently, we have developed the CVD growth of multilayer hBN to be used as a 2D insulator. We found that Fe-Ni alloy gives relatively uniform multilayer hBN and the hBN can strongly enhance the PL from WS_2 [6]. Moreover, the unique crystallographic change of the Fe-Ni thin film during the hBN growth was observed [7]. More recently, we investigated the CVD growth and transfer method of multilayer hBN and demonstrated that the CVD-grown multilayer hBN can increase the carrier mobility of monolayer graphene, as shown in Figure 1 [8].

I will also introduce our new group research project, "Science of 2.5 Dimensional Materials: Paradigm Shift of Materials Science Toward Future Social Innovation" supported by Ministry of Education, Science and Technology (MEXT), Japan that started from September 2021 [9,10].

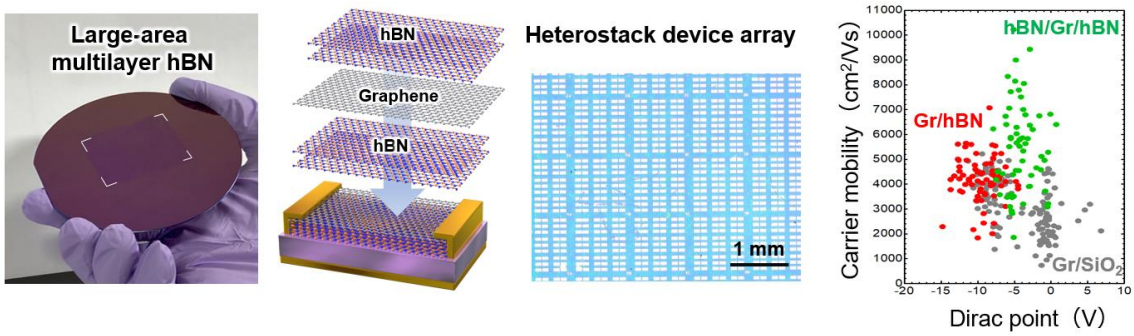

Figure 1. CVD growth of large-area multilayer hBN and application to graphene FET array [8].

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Nanofluidics-next frontiers with hBN

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In this talk, I will introduce a novel method based on liquid-activated quantum emission from native hBN defects for nanofluidic sensing. Liquids confined down to the atomic scale can show radically new properties. However, only indirect and ensemble measurements operate in such extreme confinement, calling for novel optical approaches enabling direct imaging at the molecular level. Using our method, we harness quantum emission originating from native defects in hexagonal boron nitride (hBN) for molecular imaging and sensing in nanometrically confined liquids. We show that defect activation occurs through chemisorption of organic solvent molecules, revealing single-molecule dynamics at the interface through spatially correlated activation of neighboring defects. Defect emission spectra further offer a direct readout of local dielectric properties, unveiling increasing dielectric order under nanometer-scale confinement. Liquid-activated native hBN defects bridge the gap between solid-state nanophotonics and nanofluidics, opening new avenues for nanoscale sensing and optofluidics.

https://arxiv.org/abs/2204.06287

Growth of suspended hexagonal boron nitride on GaN substrate by MOCVD

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Hexagonal boron nitride (h-BN), an insulating two-dimensional layered material, has recently attracted a great attention due to its fascinating optical, electrical, and thermal properties, and promising applications across the fields of photonics, quantum optics, and electronics. However, mechanically exfoliated bulk h-BN and h-BN films grown on catalytic metal substrates have been mainly used to study the fundamental properties, lacking in scalability for practical implementation of h-BN.

Here, we exploit the scalable approach to grow h-BN on epitaxial gallium nitride (GaN) substrate by using metal-organic chemical vapor deposition (MOCVD). It was found that at a specific MOCVD growth condition, a very unique h-BN film can be grown on GaN substrates, in which few-layer h-BN film is suspended on GaN nanoneedles. The combination of state-of-the-art microscopic and spectroscopic analyses revealed that the suspended h-BN films exhibit unprecedented DUV photoluminescence spectra with local variation in band-edge emission. In addition, the h-BN films show unprecedented atomic stacking configuration, the mechanism of which will be discussed with optical and structural characterizations and theoretical calculations.

Hexagonal boron nitride crystal growth from molten metal solutions

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Molten metal solution growth of hexagonal boron nitride is a versatile process that produces high quality crystals. First, a boron source (elemental boron or hBN) is dissolved into molten metals under nitrogen at 1550 °C. Nickel, iron, and cobalt are all good solvents for boron, while alloying with chromium increases the nitrogen solubility. Next, slow cooling (1 to 4 °C/hour) causes the solution to supersaturate and precipitate the hBN crystals on the surface of the metal. The crystals thus formed exhibit intense Cathodoluminescent and photoluminescent peaks above 5.75 eV and narrow Raman peak widths, 7 cm^{-1} at 1365.6 cm^{-1}. Boron isotope-enriched hBN crystals including $h^{10}BN$ and $h^{11}BN$ have been grown using isotopically enriched elemental ^{10}B and ^{11}B and natural nitrogen (>99% ^{14}N). Monoisotopic hBN crystals, those containing only one boron isotope instead of the 20% ^{10}B and 80% ^{11}B present in natural boron, have reduced isotopic disorder, and thus longer phonon lifetimes and higher thermal conductivity. In addition, the mechanical, thermal, and optical properties of the material also vary with the isotope concentrations. Carbon doping was demonstrated by adding carbon to the solution; this produced regions of the Bernal BN polytype crystals, which have potential application in short-wavelength optoelectronics. Cathodoluminescence studies are ongoing to determine the defects responsible for signature spectral features. Future research plans are to scale the equipment to grow larger area crystals, which is possible because the process is conducted at atmospheric pressure.

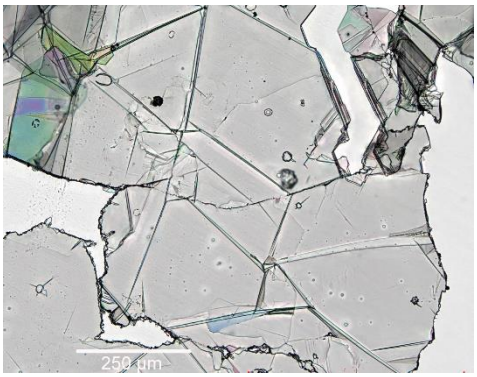

Figure 1 Optical micrograph of a representative 17 µm thick hBN crystal precipitated from a nickel-chromium solution.

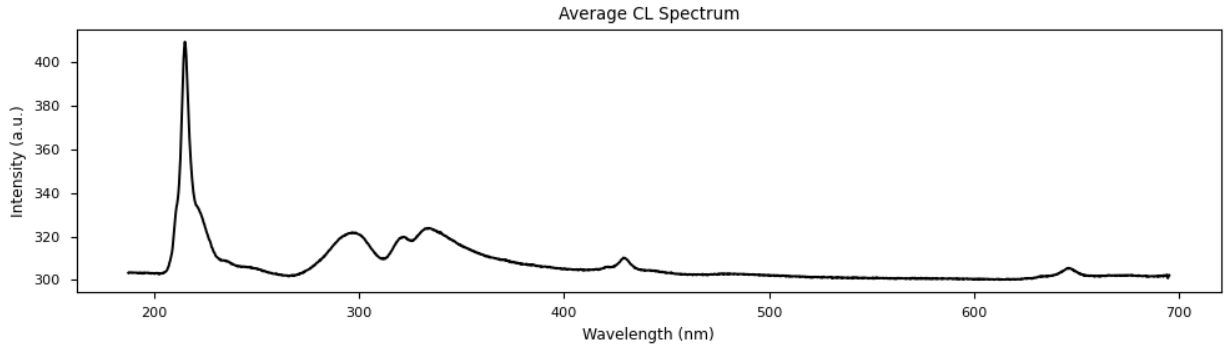

Figure 2 Cathodoluminescence spectra from an hBN crystal grown from a Co-Cr solution. The intense band-edge peak compared to the defect peaks indicate the crystal's high quality.

Molecular Beam Epitaxial Growth of Monolayer and Multi-layer h-BN

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Monolayer hexagonal boron nitride (hBN) has been widely considered as a fundamental building block for two-dimensional (2D) heterostructures and devices. However, the controlled and scalable synthesis of hBN and its 2D heterostructures has remained a daunting challenge. In this context, we have studied the epitaxy of h-BN by using ultrahigh temperature (up to 1800 °C) plasma-assisted molecular beam epitaxy (MBE) on sapphire, Ni and HOPG substrates. We show that, when grown at sufficiently high temperature, h-BN can exhibit predominantly excitonic emission at ~220 nm. The measured luminescence intensity is significantly higher than AlN under identical conditions. Moreover, we present a hBN/graphene (hBN/G) interface-mediated growth process for the controlled synthesis of high-quality monolayer hBN. We show that the in-plane hBN/G interface can be precisely controlled, enabling the scalable epitaxy of unidirectional monolayer hBN on graphene, which exhibits a uniform moiré superlattice consistent with single-domain hBN, aligned to the underlying graphene lattice. By exploiting strong excitonic effects and efficient exciton-phonon coupling, it is further observed that an indirect bandgap semiconductor such as h-BN can be transformed to be a high efficiency far UV-C light emitter. The room temperature internal quantum efficiency is measured to be 60% in the far UV-C (220 nm), which is orders of magnitude higher than that of any other known indirect bandgap semiconductors. This work provides a viable path for the controlled synthesis of ultraclean, wafer-scale, atomically ordered 2D quantum materials, as well as the fabrication of 2D quantum electronic and deep UV optoelectronic devices.

Toward coherent single photon emission from hexagonal boron nitride

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Solid state quantum light sources are emerging as promising candidates for applications in various quantum technologies. Among these sources, optically active point defects in hexagonal boron nitride (hBN) are attracting considerable attention due to their extreme brightness, high Debye Waller factor and robustness. In addition, numerous recent studies have shown that several defects in hBN exhibit spin dependent optical transitions which can be read out by optically detected magnetic resonance (ODMR) technique. This property is vital for employment of hBn defects as solid-state qubits and quantum sensors. In this talk, a summary of recent works on study of these emitters will be presented. Specifically, cryogenic spectroscopy of single defect in hBN and in-depth investigation of their optical coherence will be covered. Fabrication and optical properties of stable blue emitters in hBN will be presented and their stable photoluminescence and coherence will be explained in terms of permanent optical dipole moment and symmetry. On the rest of the talk I will cover our progress on fabrication of optical cavities from hBN with high quality factor at wavelength as low as 400 nm. Finally, our attempt of isolating single spin defect in hBN through coupling them to plasmonic and dielectric cavities will be covered.

Protection of the Spin Coherence of Defects in Hexagonal Boron Nitride

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Hexagonal boron nitride (hBN) provides a potential platform for quantum photonic technologies, thanks to the potential for its photonic integration, along with defects with appealing optical properties and optically addressable spin-levels. The negatively charged boron vacancy (V_B^-) is the most well-studied spin defect in hBN and, so far, the only one to be identified with any degree of certainty.

The 2D nature of hBN makes it an attractive platform for sensing, as the probe can be placed in close proximity to the target. However, the nuclear spin environment limits the spin echo coherence times to $\backsim$100 ns at room temperature accessible magnetic fields. To overcome this, we introduce a method to improve the spin coherence time of V_B^- ensembles. A strong modulated microwave drive is used to stabilize the Rabi oscillation, extending the coherence time up to 4 μs, which is an improvement of ~150 times and close to the 10 μs electron spin lifetime in our sample. This approach is used to define a protected qubit basis. We show full control of this protected qubit and measure a coherence time of a superposition of the protected qubit of 0.8 μs [1].

Furthermore, we apply this continuous dynamic decoupling scheme to AC magnetometry. We measure a sensitivity of $\sim 1\mu\mathrm{T}/\sqrt{\mathrm{Hz}}$ for a signal at $\sim 4\ \mathrm{GHz}$ and demonstrate that the microwave control signal can be used to tune the sensitivity, providing a means to determine the signal's frequency, with a bandwidth of $\sim 300\ \mathrm{MHz}$ for a fixed DC magnetic field.

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Defect spins and qubits in hexagonal boron nitride from first principles theory guiding experiments

Song Li, Anton Pershin, Péter Udvarhelyi, Viktor Ivády and Adam Gali

We have recently developed first principles techniques for theoretical microscopy and spin control of point defects in semiconductors and insulators. We have applied these techniques to several point defects in hexagonal boron nitride (hBN) that are relevant in real hBN materials. We proposed to apply the negatively charged boron-vacancy to realize qubits. This center was indeed later observed in experiments. We have recently simulated the coherence time and Rabi-oscillation of this center with showing an entanglement between the electron spin and the first four neighbor ^{14}N spins. We have recently identified the coupling of the electric field and the strain to the electron spin in the ground state of the center with quantifying the theoretical sensitivity limits and interpreting the optically detected magnetic resonance (ODMR) spectrum of ensembles. We identified an oxygen-related electron paramagnetic resonance center in hBN which produces 2-eV emission with relatively small phonon sideband. Furthermore, we proposed two types of ultraviolet (UV) emitters in hBN: (i) the 5-7 Stone-Wales defects and (ii) the carbon-pair defect structures with the most stable form of a carbon ring. We shall show how the properties of the UV emitters change upon stacking sequences of hBN layers.

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Nuclear spin control in hexagonal boron nitride

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Electron spins in van der Waals materials are important for many recent developments in condensed matter physics and spintronics, such as topological insulators and magnetic tunnel junctions. On the other hand, nuclear spins in van der Waals materials remain largely unexplored due to the lack of suitable tools. Recently, we created negatively charged boron vacancy (V_B^-) spin defects in hexagonal boron nitride (hBN) with femtosecond laser writing [*ACS Photonics*, 8, 994 (2021)] and ion implantation. We also increased the brightness of V_B^- spin defects in hBN with plasmonic enhancement [*Nano Letters*, 21, 7708 (2021)], and optically polarized nitrogen-14 nuclear spins in hBN with the help of V_B^- electron spins [*Nature Materials* 21, 1024 (2022)]. In addition, we observed Rabi oscillation of nuclear spins in hBN and strong electron-mediated nuclear-nuclear spin coupling. Our work opens new avenues for the manipulation of nuclear spins in van der Waals materials for quantum information science and technology.

Quantum sensing and imaging with spin defects in hBN

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Abstract:

Hexagonal boron nitride (hBN) has recently emerged as a promising material for realising ultrathin quantum sensors based on optically addressable spin defects. In particular, two types of spin defects have been reported, the well-studied boron vacancy defect emitting in the near infrared, and a family of defects emitting primarily in the visible. In this talk, I will discuss our recent work on developing these systems for quantum sensing and imaging. First, I will present experiments where boron vacancy defects in exfoliated hBN flakes are used to image static magnetic fields from ferromagnets and electric currents, as well as to image temperature distributions. I will then discuss how hBN nanopowders containing both boron vacancies and visible-emitting spin defects can be used as a versatile platform for sensing and imaging. The advantages and limitations of hBN for quantum sensing compared to established systems like the NV centre in diamond will be discussed.

Coherent Control and Sensing Applications of the Boron Vacancy in hexagonal Boron Nitride

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Van der Waals (vdW) materials have emerged over the last decade as the new playground for quantum photonics devices. Among them, hexagonal boron nitride (hBN) is an interesting candidate, mainly because of its crystallographic compatibility with many different 2D materials, but also because of its ability to host optically active spin defects. We have recently reported [1] the optically detected magnetic resonance of spin-triplet negatively charged boron vacancies (V_B^-) in hBN and determined their spin-Hamiltonian parameters. Furthermore, we demonstrated the coherent control of VB^- at room temperature and determined the relevant spin-coherence times. [2]

In this respect, sensors based on such colour centres embedded in an intercalated hBN layer within a vdW heterostructure may be particularly attractive, since the distance between the sensor and the object to be sensed can be quite small. The influence of external stimuli (magnetic field, temperature, pressure, etc.) on this spin defect will be also discussed. [3]

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Mass-production of two-dimensional h-BN and its liquid crystals for deep UV light modulation

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Light modulation is important for optical devices. For example, liquid crystal based display is a typical light modulation technology with a global annual market over 100 billion US$. Nevertheless, modulation of light at extreme wavelengths like deep UV (wavelength <300 nm) is challenging despite its importance in many applications. Current technologies cannot do it because the widely-used organic liquid crystal molecules are not stable under deep UV light, and birefringence of inorganic crystals is difficult to be tuned by external fields. In this talk, I will discuss our recent work on the fabrication of a stable and tunable deep UV light modulator by using 2D h-BN.[1-2] The ultra-wide optical bandgap, high stability in deep UV, and large optical anisotropy factor resultant sensitive magnetic-field response, collectively making 2D h-BN an ideal candidate for stable and continuous deep UV light modulation. Such 2D h-BN can be produced in large quantities by scaled top-down exfoliation technologies,[3-4] suggesting its clear potential for practical uses.

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Boron nitride nanotube and nanosheet properties and functions as revealed by *in situ* transmission electron microscopy

Electrical and mechanical properties of pristine and carbon-doped BN nanotubes[1] and graphene-like nanosheets, so called "boronitrines", are investigated using state-of-the-art methods of *in situ* and *operando* high-resolution transmission electron microscopy (TEM). Elastic modulus, tensile strength,[2,3] fracture toughness, conductivity of 1D and 2D BN-based nanostructures are measured in different morphologies, dimensions and defectiveness of the objects. Various modern *in situ* holders allowing for current-voltage and/or force-displacement curve measurements and shining light of various wavelengths onto the nanostructures are utilised in high-resolution electron microscope columns under atomic resolution. Based on the TEM results, emerging practical applications of BN-nanomaterials as effective supports for most common catalytic reactions, *e.g.*, CO_2 hydrogenation, CO oxidation *etc*., hydrogen accumulation,[4] secondary-ion batteries, reinforcing agents in light metals and polymers, and water purifiers are also shown. The results are supported by first-principle theoretical calculations.

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Ladder Ferroelectricity

Moshe Ben Shalom

We demonstrate and study a polar layered system with distinct ladder-like polarization steps that accumulate with each extra atomic layer. Moreover, the symmetries of these diatomic crystals translate planar shifts by one interatomic spacing to the out-of-plane switching of the structure and its polarization. I will discuss the origin of this ultimately thin interfacial polarization, the unique cumulative response at the atomic limit, the robust co-existence with in-plane conductivity, and the switching dynamics observed in our experiments and modeled by our first principle calculations.

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https://www.nature.com/articles/s41586-022-05341-5
https://arxiv.org/abs/2206.12215

Current status and challenges in hBN growth by chemical vapor deposition

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Hexagonal boron nitride (hBN) is a promising two-dimensional (2D) material owing to its unique optical properties in the deep-UV region, mechanical robustness, thermal stability, and chemical inertness. hBN thin films have gained significant attention for various applications, including nanoelectronics, photonics, single photon emission, anti-corrosion, and membranes. Thus, wafer-scale growth of hBN films is crucial to enable their industrial-scale applications. In this regard, chemical vapor deposition (CVD) is a promising method for scalable high-quality films. To date, considerable efforts have been made to develop continuous hBN thin films with high crystallinity, from those with large grains to single-crystal ones, and to realize thickness control of hBN films by CVD. However, the growth of wafer-scale high crystalline hBN films with precise thickness control has not been reported yet. The hBN growth is significantly affected by substrate, in particular the type of metals, because the intrinsic solubilities of boron and nitrogen depend on the type of metal. In this talk, state-of-the-art strategies adopted for growing wafer-scale, highly crystalline hBN are summarized, followed by the proposed mechanisms of hBN growth on catalytic substrates. Furthermore, various applications of the hBN thin films are demonstrated, including a dielectric layer, an encapsulation layer, a wrapping layer of gold nanoparticles for surface enhanced Raman scattering, a proton-exchange membrane, a template for growth of other 2D materials or nanomaterials, and a platform of fabricating in-plane heterostructures. Finally, the inherent challenges are summarized, and future research directions for the facile CVD-based growth of single-crystal hBN are proposed.

High-temperature MBE of hBN monolayers and graphene-hBN lateral heterostructures.

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High-temperature molecular beam epitaxy (HT-MBE) approach offers new opportunities for the growth of graphene and hBN two-dimensional (2D) materials and related van der Waals (vdW) heterostructures.

We have developed HT-MBE growth of hBN at growth temperatures from 1250ºC to 1700ºC using sublimation and e-beam boron sources. By growing hBN on highly oriented pyrolytic graphite (HOPG) substrates, we have produced monolayer and few-layer thick boron nitride with atomically flat hBN surfaces, which are essential for 2D and DUV applications. The hBN coverage can be reproducibly controlled by the growth time, substrate temperature and boron to nitrogen flux ratios. We will discuss our recent measurements of a direct optical energy gap of ~6.1 eV [1] and electronic band gap of ~6.8 eV [2] in single monolayer hBN. We will present data demonstrating that the single-photon emitters (SPEs) in hBN are related to carbon (C) incorporation [3] and will discuss C-doping techniques in HT-MBE.

We have used different types of the MBE carbon sources, including a sublimation source, atomic carbon source and e-beam source, to grow graphene on sapphire, SiC and hBN substrates at temperatures between 1000ºC and 1700ºC.

We have demonstrated that lateral heterojunctions of graphene and hBN can be grown *in-situ* using HT-MBE, including epitaxy of monolayer-thick lateral hBN-graphene-hBN heterostructures, in which a strip of graphene is embedded between hBN monolayers.

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Wafer-scale (MO)CVD Synthesis of Hexagonal Boron Nitride and Graphene on Sapphire

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Two-dimensional (2D) materials have been identified as key enablers for future logic applications while (metal-organic) chemical vapor deposition (MO(CVD)) technique is the first choice of the industry to implementing 2D material synthesis on a wafer-scale [1]. In this talk we present results of wafer scale growth and characterisation of graphene, hexagonal boron nitride (hBN) and their heterostructures on sapphire substrate. Both graphene and hBN synthesis is achieved in a commercial AIXTRON Close Couple Showerhead (MO)CVD reactor reaching substrate temperatures up to 1400 °C and accommodating wafers up to 200 mm in diameter [2, 3]. Such setting allows highly uniform deposition across the wafer exemplified by near-field terahertz (THz) and Raman spectroscopy of a single-layer graphene film (**Figure 1 a, b**) grown via CVD using methane as a precursor. For hBN, both CVD using borazine precursor and (MO)CVD route using triethylborane and ammonia have been explored. The hBN film deposition was scaled-up to a wafer diameter of 200mm (**Figure 1c**). Film quality was assessed by X-Ray reflectivity indicating 9 nm thick film with less than 0.7 nm film thickness variation across the wafer. A blue-shift of the hBN Raman peak towards the center (**Figure 1d**) indicates compressive stress. Further, growth of hBN/graphene heterostructure in-situ has been successfully demonstrated. Finally, single layers of graphene and hBN were transferred to characterize their electrical properties. Charge carrier mobility of over 1400 cm^2/Vs was demonstrated in a transferred graphene layer, while resistive-switching of hBN sandwiched between metal electrodes was explored for memristor applications.

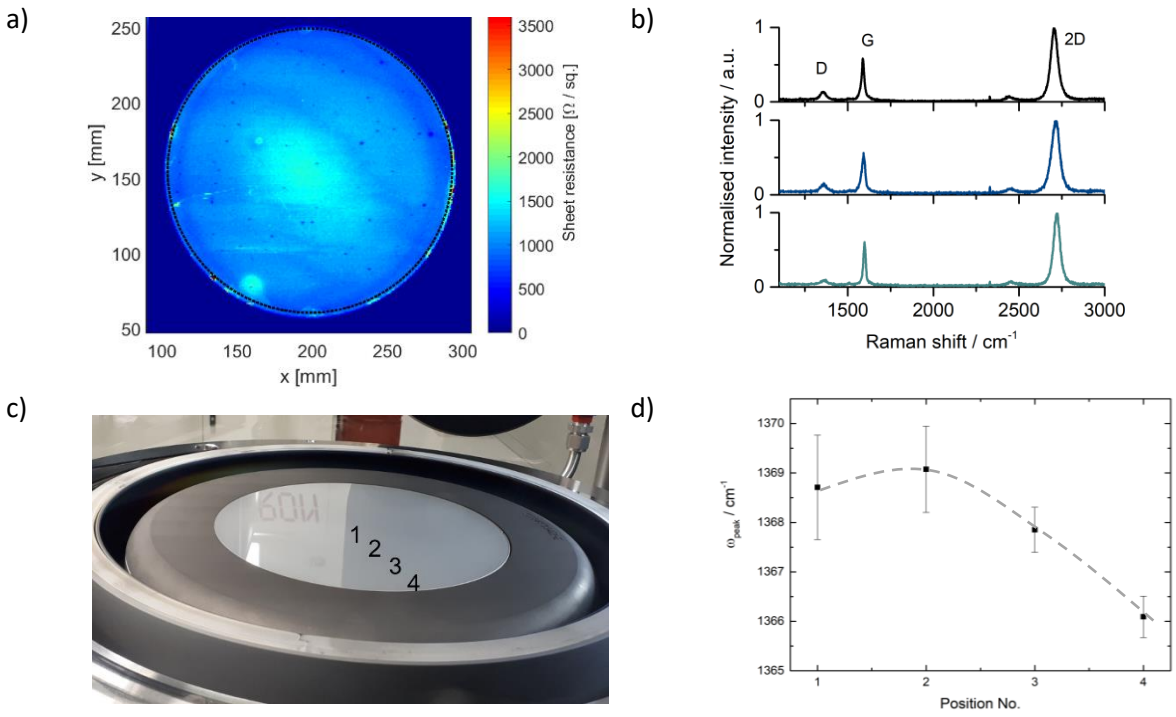

Figure 1. a) Sheet resistance mapping of 200 mm graphene/sapphire wafer characterised by THz spectroscopy; **b)** corresponding Raman spectra at different locations across the wafer. **c)** hBN on 200mm sapphire; **d)** peak position of hBN Raman signal at various positions indicated in c).

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Abstract: Select quantum materials can support polaritons, which are hybrid light matter waves with sub-diffraction-limited confinement. In this talk, I will discuss polaritons in hyperbolic materials, which propagate as conical rays throughout the bulk of these crystals. I will introduce polaritons in hyperbolic hetero-bicrystals. Our data reveals negative refraction, spectral gaps, and wave localization in a hetero-bicrystal made of the two thin crystals: Molybdenum oxide and isotopically pure hexagonal boron nitride [1]. Some of the challenges and opportunities for polaritons in quantum materials, including hetero-bicrystals, will be discussed.

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Molecules-BN interaction via polaritons

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Phonon polaritons – lattice vibrations coupled to electromagnetic fields – in van der Waals (vdW) materials can enhance light–matter interactions at mid-infrared frequencies, owing to their extreme field confinement and long lifetimes. Particularly, in h-BN the dispersion of polaritons – the relation between the momentum and energy – can take a hyperbolic shape and lead to the strong coupling between the polaritonic fields and molecular vibrations. In this talk we demonstrate that vibrational strong coupling can be achieved between phonon polaritons either freely propagating along h-BN slabs [1] or "locked" inside resonant cavities [2] and molecular vibrations in adjacent thin molecular layers. Such interaction can take place simultaneously in different frequency bands, e.g. at visible and mid-infrared frequencies [3]. We will show the most recent experimental and theoretical studies on the interaction between hyperbolic polaritons and molecules, discuss their applications and future perspectives.

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Figures

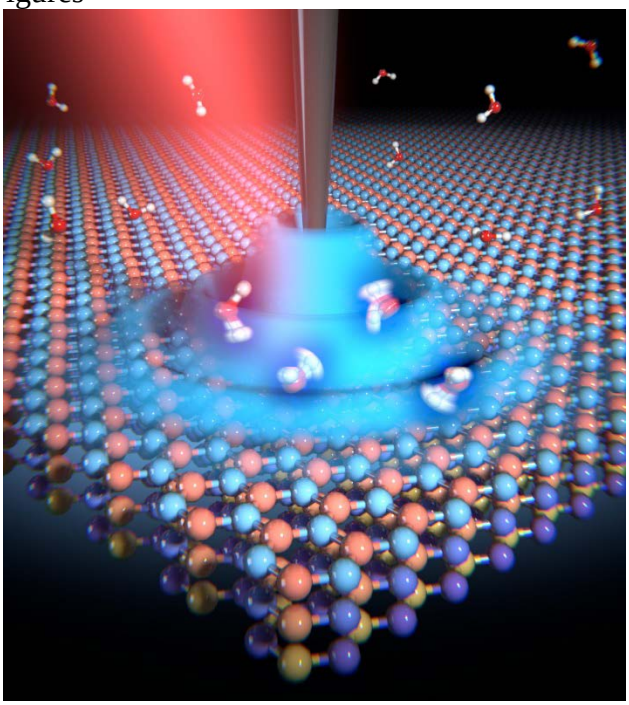

Figure 1: An artistic image of a phonon polariton in h-BN interacting with molecular vibrational resonances. The vertical rod represents the AFM tip of a near-field optical microscope, probing the interaction.

Hexagonal Boron Nitride as a Low-loss Dielectric for High-performance, Small-footprint Superconducting Qubit Devices

Dielectrics with low loss at microwave frequencies are imperative for high-coherence solid-state quantum computing platforms. Owing to its chemical stability and atomic flatness, hexagonal boron nitride (hBN) may enable high-quality superconducting devices such as parallel-plate capacitors (PPC) and Josephson junctions for building superconducting quantum circuits with small form factors.

We study the dielectric loss of hexagonal boron nitride (hBN) thin films in the microwave regime by measuring the quality factor of parallel-plate capacitors (PPCs) made of $NbSe_2$-hBN-$NbSe_2$ heterostructures integrated into superconducting circuits. The extracted microwave loss tangent of hBN is bounded to be at most in the mid-10^{-6} range in the low-temperature, single-photon regime.

The hBN PPC is further integrated with aluminum Josephson junctions to realize transmon qubits with coherence times reaching 25 μs, consistent with the hBN loss tangent inferred from resonator measurements. The hBN PPC reduces the qubit feature size by at least two orders of magnitude compared to conventional all-aluminum coplanar transmons. The high confinement of the electric field (up to 91%) in the hBN region may also reduce unwanted qubit cross-talk. Finally, I will discuss Josephson junction devices using thin hBN (2L~6L) tunneling barriers and how these hBN heterostructures may lead to all-vdW, merged-element superconducting qubits.

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Wang, J.IJ., Yamoah, M.A., Li, Q. *et al.* Hexagonal boron nitride as a low-loss dielectric for superconducting quantum circuits and qubits. *Nat. Mater.* **21**, 398–403 (2022). https://doi.org/10.1038/s41563-021-01187-w

First observation of bernal boron nitride single crystals

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Hexagonal boron nitride is a wide band gap semiconductor which differs from the other nitrides because of its sp2 hybridized bonds forming honeycomb atomic planes weakly stacked through van-der-Waals interactions [1]. Since the first synthesis of large hBN single crystal by Watanabe and Tanigushi in 2004 [2], hBN has become a pivotal crystal in a wide variety of applications [1]. However, even though the hBN polytype (namely the AA' stacking) is the most commonly observed in high quality crystals, other boron nitride sp2 polytypes with different stacking can be formed [3, 4]. The calculations made on these polytypes predict changes of the band gap properties [5, 6]. Until now, no boron nitride sp2 polytypes single crystal other than hBN has been observed and little is known about their properties.

In this work we report the first observation of a bernal boron nitride high quality single crystal. In contrast to hBN which is centrosymmetric prohibiting second harmonic generation (SHG), the bernal polytype is non-centrosymmetric and SHG can be observed. By combining hyperspectral photoluminescence and SHG spectroscopy and photoluminescence excitation (PLE) spectroscopy on carbon-doped sp2 boron nitride crystals synthetized by the Ni-Cr flux method we identify a 10um size bernal boron nitride single crystal between two hBN single crystals. We then unravel the optical properties in correlation with calculations, where indirect and direct bandgaps are quasi-degenerate. This work paves the way for research on sp2 boron nitride polytype and their potential applications in optoelectronics.

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Probing deep-ultraviolet optoelectronic processes in hexagonal boron nitride

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Hexagonal boron nitride (hBN) is a van der Waals (vdW) semiconductor with a wide bandgap of ~ 5.96 eV. Despite the indirect bandgap characteristics of hBN, charge carriers excited by high energy electrons or photons efficiently emit luminescence at deep-ultraviolet (DUV) frequencies via strong electron-phonon interaction. In this work, we probe optoelectronic processes at a band edge in hBN by means of optical imaging and spectroscopy at deep ultraviolet frequencies. Our laser excitation spectroscopy shows that strong radiative recombination and carrier excitation processes originate from the pristine structure and the stacking faults in hBN. We further demonstrate prominent electroluminescence and photocurrent generation from hBN by fabricating vdW heterostructures with graphene electrodes. Our work provides a pathway toward efficient DUV light emitting and detection devices based on hBN.

P/N Type Conductions and Large-Scale Synthesis of Hexagonal Boron Nitride

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Hexagonal boron nitride (h-BN) suffers lots of fundamental difficulties, including large-scale preparation, controllable heteroepitaxy on various substrate, and efficient p and n type conductivities for optoelectronic applications. Here, we summarize our recent work on advances of 2D h-BN films.

1) By winding the Cu foil substrate into mainspring shape supported by a multi-prong quartz fork, an extremely large-size monolayer h-BN film has been achieved over 25 inches in a 1.2-inch reactor.

2) h-BN film was explored for van der Waals heteroepitaxy of GaN/ AlN and preorientated growht of ZnO nanorod array. It has been transferred onto 2"/4" wafer as a release buffer layer. Overgrowth of thick GaN over 200 μm has been achieved free of residual strain. On polycrystalline Cu substrate, highly aligned ZnO nanorods array (~ 75 μm) was preoriented by h-BN monolayer. A ultrahigh responsitivity solar-blind photodetecting paper has been fabricted (7000 A/W @ 265 nm).

3) By orbital engineering, both *p*- and *n*-type conductivities in h-BN monolayer have been obtained for the first time. The efficient *p*-type conduction in h-BN was obtained via Mg *s*-orbital modulated doping and the hole current can reach 32 μA. A novel strategy of orbital level engineering through sacrificial impurity coupling between Ge-O was proposed to achive *n*-type conductivity with a considerable current of ~100 nA.

It is believed that 2D h-BN semiconducting layer will lead to broad applications in various advanced optoelectronic devices.

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Contributed

Far/Mid ultraviolet electroluminescence from an electrically induced color center in hexagonal boron nitride

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Hexagonal boron nitride (hBN) is a two-dimensional van der Waals material composed of boron and nitrogen atoms in a hexagonal lattice. hBN is the wide-bandgap semiconductor with a band of 6.4 eV and shows efficient band edge cathodoluminescence at 215 nm and lasing behavior. Here I will present the efficient Far/Mid UV electroluminescence (EL) in band edge emission at 215 nm as well as broad 303-333 nm emission peaks from the hBN van der Waals heterostructure. We observed that 303-333 nm broad EL emissions with phonon replica of optical phonon energy of hBN based on the Franck-Condon principle, which are attributed to the local electric field induced color centers and its highly localized excitonic features. I will also present the tunable color center emission as a function of electric field direction and discuss the possible origin of color centers in hBN van der Waals heterostructures. These results demonstrate the promising developments of a highly efficient solid-state DUV light source and solid-state quantum emitter at the nanoscale and allow the development of the key architectures for the DUV nanophotonics, bio-sensing, high-precision metrology, and quantum information.

ATOMIC SCALE MAPPING OF THE ELECTRIC FIELD AND CHARGE DENSITY IN BN NANO-STRUCTURES BY 4D-STEM

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Atomic scale changes of the electric field in 1D and 2D materials play a key role in defining physical and chemical properties such as reactivity, interface effects and molecule rearrangement. However, imaging electric field variations at the individual atom sensitivity and space resolution is still a challenge.

In this regard, four-dimensional scanning transmission electron microscopy imaging (4D-STEM) has recently appeared as a very promising technique. In 4D-STEM experiments, a 2D convergent beam electron diffraction pattern is acquired at each probe position, generating a 4D data set. The displacements of the center of mass (COM) of the diffraction pattern is directly related to the interaction between the electron beam and the electrostatic field during its propagation through the material. In the case of weak phase objects, this technique is in principle quantitative and give access to spatial variations of electric fields down to the atomic scale.

In this work we present 4D-STEM analysis performed on perfect and defective mono- to few-layers h-BN flakes and compare them with simulations obtained using the Coulomb potential derived from full-potential density functional theory calculations. We demonstrate that the method is fully quantitative when both electronic and nuclei charges are taken into account. We further discuss how very high COM displacements at sample discontinuities may be wrongly attributed to strong enhancing of the electric field while they derive to probe size effects.

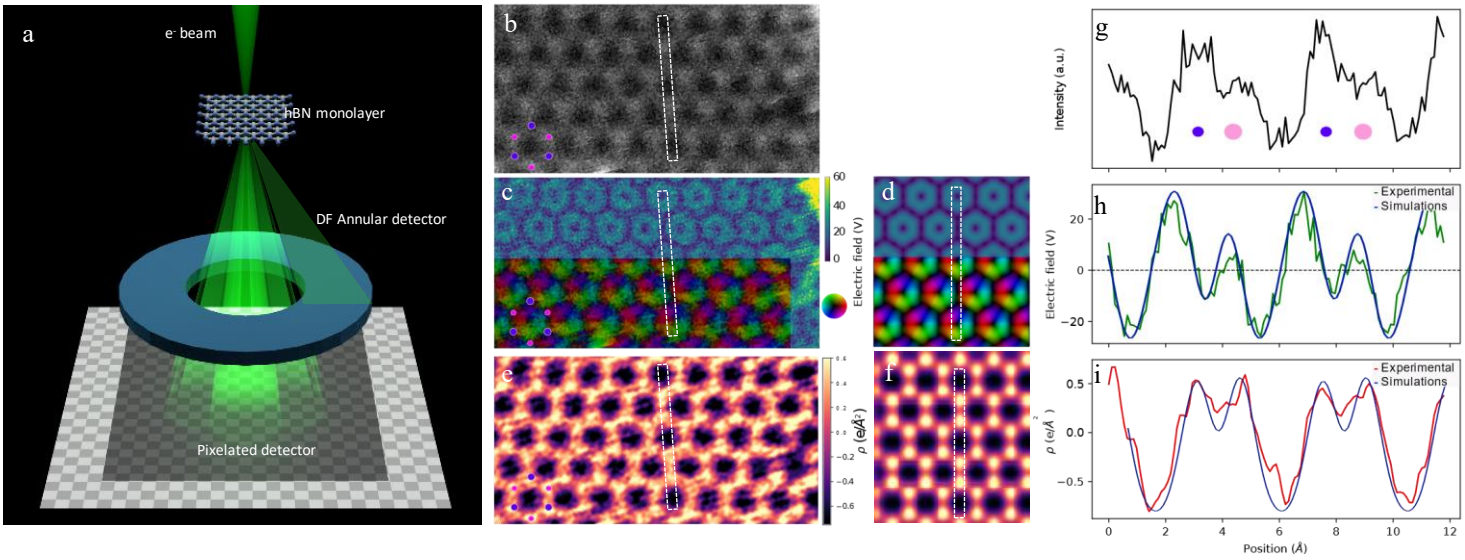

Figure 1: (a) 4D-STEM measurements geometry (b) ADF image and (c) electric field intensity and orientation map of h-BN monolayer compared with the electric field obtained by multi-slice simulations (d). (e) Experimental and (f)simulated charge density map. (g,h,i) Intensity line profiles of ADF, experimental and simulated electric field and charge density taken along the indicated direction.

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Room-temperature coherent control of single carbon-related defects in hexagonal boron nitride

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Optically addressable spins in materials are important platforms for quantum technologies, such as repeaters and sensors[1]. Spins in two-dimensional layered materials offer a particular advantage, as their reduced dimensionality enables feasible on-chip integration into devices. In recent years, optically addressable spins have been identified in hexagonal Boron Nitride (hBN), attributed to ensembles of boron vacancy defects (VB^-)[2][3] and single carbon-related defects[4][5]. In this talk I will present the room-temperature optically detected magnetic resonance (ODMR) of single carbon-related spin defects as well as, most recent results of coherent control of these spin defects. These results portray the surprising spin physics at play in this 2D system with a rich nuclear-spin environment. In addition, our results show an important correlation between the bunching dynamics and presence of ODMR, and that the ODMR signatures indicate a S=1 system. Our results present a promising route towards realising a room-temperature spin-photon quantum interface in hexagonal boron nitride.

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Epitaxial Hexagonal Boron Nitride for Hydrogen Generation by Radiolysis of Interfacial Water

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Hydrogen is an important building block in global strategies toward a future green energy system. To make this transition possible, intense scientific efforts are needed, also in the field of materials science. Hexagonal boron nitride (hBN) is a very promising candidate for such applications, as it has been demonstrated that micrometer-sized flakes are excellent barriers to molecular hydrogen [1,2]. However, it remains an open question whether large-area layers fabricated by industrially relevant methods [3] preserve such promising properties.

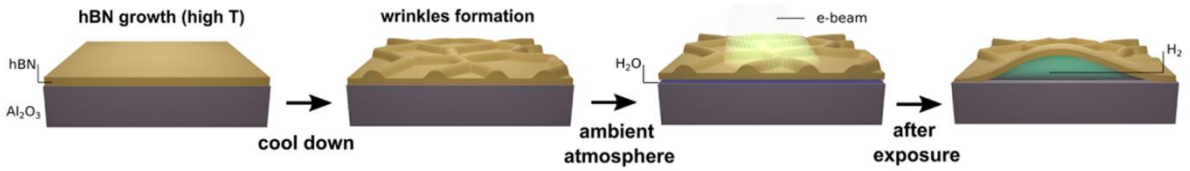

Fig. 1: Schematic illustration of the bubble formation. hBN is grown by MOVPE at temperatures above 1000 °C. After the growth the sample is cooled to room temperature, which leads to the formation of hBN wrinkles. The sample is removed from the reactor and exposed to ambient conditions. Electron beam exposure in an SEM leads to bubble formation [4].

In this work, we show that electron-beam-induced splitting of water creates hBN bubbles (see Fig. 1) that effectively store molecular hydrogen for weeks [4]. Raman spectroscopy proves the presence of molecular hydrogen and experiments with heavy water provide evidence that hydrogen generation is triggered by the radiolysis of interfacial water. By performing a stress test we could also demonstrate that H_2 remains in the bubble even after extreme wear and deformation (over 500 cycles of deflation and inflation by changing the ambient pressure), highlighting the suitability of our large-area epitaxial material for possible hydrogen storage applications.

Our findings show that hBN is not only a potential candidate for hydrogen storage but also holds promise for the development of unconventional hydrogen production schemes.

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Two-photon interference from position-controlled quantum emitters in hBN

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3 LPENS - Laboratoire de physique de l'ENS - ENS Paris

In the context of photonic quantum information science, hexagonal boron nitride (hBN) has emerged as a very promising material. The two-dimensional character of hBN renders it attractive for the realisation of compact heterostructures and integrated photonic devices. Moreover, this wide-gap material has been shown to host single-photon emitters (SPEs) with appealing optical properties in the red and near infrared regions [1]. However, the deep defects initially observed in hBN suffer from the wide distribution of their emission wavelength and, in most cases, a random spatial location [2,3]. These limitations hinder the scalability of the system for applications.

Recently, a new family of quantum emitters has been observed in hBN – a class of blue-emitting colour centres (λ ~ 435 nm) that have appealing properties [4,5,6]. We demonstrate controlled positioning of these SPEs with reproducible emission wavelengths [5,7]. The SPEs are locally activated in exfoliated hBN flakes using a focused electron beam and subsequently characterised using microphotoluminescence. They exhibit narrow linewidths at low temperature and a drastically reduced ensemble distribution of their emission wavelength ($\Delta\lambda < 1$ nm, fig. 1). Individual emitters display low $g^{(2)}(0)$ as well as high and stable count rates up to room temperature. We investigate two-photon interference between consecutively emitted photons based on the Hong-Ou-Mandel effect, leading to a sizeable degree of indistinguishability ($V_{HOM} = 0.56 \pm 0.11$) [8], which opens the way to the use of these SPEs for quantum information applications.

Our results suggest new avenues towards top-down realisation of integrated quantum optical devices based on indistinguishable single photon sources in hBN.

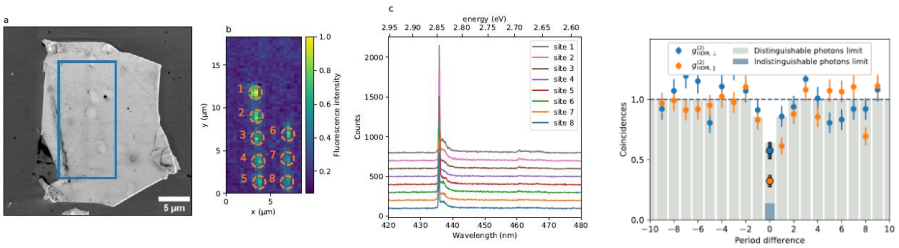

Figure: hBN flake with eight irradiation sites and corresponding confocal map and spectra, displaying reduced statistical dispersion of the emission wavelength. Right: photon correlations with a signature of Hong-Ou-Mandel effect between consecutive photons.

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Fabrication and polarization dynamics of yellow single photon emitters in Hexagonal Boron Nitride

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(Dated: February 7, 2023)

Abstract

Single-photon emitters in solid-state systems have received a lot of attention as building blocks for numerous applications in quantum technology. Atomically localized defect-based emitter in hBN stand out due to their optical and physical properties, such as room temperature operation and high single photon count rate. However, on-demand localized fabrication of these emitters in the crystal lattice is still a topic of research and due to which the integration of these emitters with scalable optical and electronic platforms remains challenging. The present work is an effort to fabricate emitters by electron irradiation with a scanning electron microscope. The sub-micron lateral precision of the SEM allows localized fabrication of these emitter in the crystal lattice. Density functional theory calculations, coupled with experimentally observed emission lines at 575 nm show that the emitters might be related to the presence of carbon-based defects. We also present results on correlating the atomic crystal structure properties and the dipole polarization dynamics. Our results presents the high yield of identical emitters, which plays crucial role for the realization of quantum applications.

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Cascade phonon polaritons in mixed-dimensional van der Waals heterostructures for broadband strong light-matter interactions

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Abstract

Phonon polaritons in polar crystals have recently gained significant attention due to their remarkable confinement and enhancement of electromagnetic fields, low group velocities, and low losses. However, these unique excitations, resulting from the coupling between photons and lattice vibrations, exhibit limited spectral responses that may limit their practical applications. Here, we propose and experimentally demonstrate that mixed-dimensional polar van der Waals heterostructures can cascade the phonon polariton responses of their polar constituents to enable broadband light-matter interactions. We transfer thin flakes of two polar van der Waals materials, hexagonal boron nitride (h-BN) and α-phase molybdenum trioxide (α-MoO3), onto a polar quartz substrate to create the mixed-dimensional heterostructure. Direct infrared nanoimaging experiments show that this heterostructure supports phonon polaritons in a broadband infrared spectral range from 900 to 1700 cm^{-1}. Further, we theoretically study the potential of using phonon polaritons in the heterostructure for achieving vibrational strong-coupling with multiple molecular absorption modes. We investigate the evolution of vibrational strong-coupling as a function of heterostructure thickness, providing the foundation for designing phonon-polaritonic biosensors based on the heterostructures. Our findings suggest that cascade phonon polariton responses in mixed-dimensional van der Waals heterostructures have the potential to pave the way for the development of broadband and integrated infrared devices for molecular sensing, signal processing, and energy control.

DETERMINISTIC CREATION AND CHARACTERIZATION OF NANOPORES IN HEXAGONAL BORON NITRIDE VIA ABERATION CORRECTED SCANNING TRANSMISSION ELECTRON MICROSCOPY (AC-STEM) AND OPTICAL MICROSCOPY

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The discovery of room-temperature single photon emission (SPE) in hexagonal boron nitride (hBN) launched it to the forefront of research as a promising platform for room-temperature quantum optics and photonics. Room-temperature quantum emission centers in hBN exhibit single-photon emission and optically addressable spin states, as desired for many quantum technologies. In this work, we create and characterize nanopores in hexagonal boron nitride (hBN) via aberration corrected scanning transmission electron microscopy (AC-STEM) drilling and photoluminescent (PL) spectroscopy. Using a finely tuned electron beam, defects on the sub-nanometer to nanometer scale are induced in hBN through electron irradiation and AC-STEM drilling. Atomic-resolution electron energy loss spectroscopy (EELS) is used to monitor and characterize the generation of these defects in real time, and atomic AC-STEM imaging is used to understand the atomic structure of the defects. Confocal PL spectroscopy is subsequently employed to characterize the optical activity of the defects created through electron irradiation and drilling. Through this work, we demonstrate the proof of principle to create defects in hBN of desired size, shape, and location. This work paves the way for controllable defect engineering in hBN and other 2D materials through AC-STEM drilling.

Posters

Optimized Irradiation Protocol for Quantum Sensors in Hexagonal Boron Nitride

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Color centers in solid-state materials show great potential in quantum information technology and sensing applications. The lately discovered negatively charged boron vacancy (V_B^-) in hexagonal boron nitride (hBN) [1] has shown that the defect exhibits a spin-triplet ground state with spin-dependent photoluminescence which enables for manipulation and read-out using optically detected magnetic resonance (ODMR). The system can be exploited in terms of its application as temperature, magnetic field, and pressure sensor [2,3], extending the already known applications of e.g., NV^- centers in diamond not only due to its 2D character but also by highly improved temperature sensing at low temperatures below 50K. Here we present an optimized irradiation protocol for the creation of V_B^- in hBN by nitrogen ions improving quantum metrology limits. We also present tremendous improvement of ODMR contrast showing hyperfine interaction on flakes of down to 80nm thickness.

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Exciton Dynamics in APHT-Grown hBN Crystals Probed by Time-Resolved Cathodoluminescence

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ABSTRACT

High quality hBN single crystals are needed for the assembly of van der Waals heterostructures. Today graphene and TMDs devices are mostly assembled using hBN grown at high pressure high temperature (HPHT) provided worlwide by a single japanese lab [1]. The high-quality HPHT crystals are grown submillimiter, their size being limited by the anvil cells needed to reach high pressures. The atmospheric pressure high temperature (APHT) method [2,3] is an alternative and potentially up-scalable method which has already proven to give hBN single crystals of sufficiently high quality for graphene electronics [4].

In this work, we implemented the APHT method using NiCr metallic solvent for hBN growth. We have fabricated centimeter scale hBN crystals, almost single crystalline as demonstrated by electron backscattered diffraction. The quality of the hBN crystal was evaluated using time-resolved cathodoluminescence at room temperature, and a free exciton lifetime of 3.2 ns (std. dev. 12%) was observed, which is comparable to the best reported values for HPHT crystals (4.2 ns) [5]. Our results demonstrate the potential of the APHT method for producing high-quality hBN single crystals, with dimensions closer to microelectronic standards.

On the APHT hBN crystals, we have performed temperature- and time-dependent measurements of exciton recombination in the temperature range of 5-300K and discuss the evolution of their exciton lifetime. We also monitored the evolution of a new color center emitting at 271 nm in the UV range with increasing electron dose. These results could help in optimizing hBN-based optoelectronic devices.

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Extending the coherence of spin qubits in hexagonal boron nitride by materials engineering: a cluster expansion theory

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Negatively charged boron vacancy (V_B^-) in hexagonal boron nitride (h-BN) has recently emerged as a new promising spin qubit candidate in 2-dimensional materials hosts for solid-state quantum applications. Their spin coherence time (T_2), however, was measured to be very short limited to a few microseconds, which is mainly due to their interaction to the inherent dense nuclear spin bath of the h-BN host. In this study, we theoretically propose ways to enhance the quantum coherence of the V_B^- spin in h-BN by using isotopic and strain engineering. We combine density functional theory and cluster correlation expansion to compute the decoherence of V_B^- spins induced by the dense nuclear spin bath of h-BN. We show that inhomogeneous strain can create spatially varying nuclear quadrupole interaction in h-BN, which can significantly suppress the nuclear spin flip-flop dynamics in the bath. In addition, we find that the coherence time of the V_B^- spin can be effectively engineered by adjusting the ratio of ^{10}B and ^{11}B isotopes in h-BN. We show that the combination of the two methods could increase the T_2 time by 4.5 times larger than the T_2 time in a pristine h-BN bulk. Our results provide not only a fundamental understanding of the decoherence of V_B^- spins in h-BN, but pave the way to engineer their T_2 time, which is crucial for their practical applications.

INVESTIGATION OF THE MATERIAL PROPERTIES OF AMORPHOUS BORON NITRIDE

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Abstract

Amorphous boron nitride (aBN) has attracted significant attention as an ultralow dielectric constant material ($\kappa<2$) with strong thermal stability and mechanical properties, making it highly suited for next generation interconnect technologies [1,2]. The structure of amorphous materials depends strongly on the fabrication process, which allows the tuning of specific properties for applications. Therefore, new fabrication strategies to control these structural properties and a characterization of their impact on the thermal, mechanical and electronic properties are urgent.

In this work, we experimentally and theoretically investigate the thermal and mechanical properties of aBN as a function of external parameters such as temperature, quenching rate, or the presence of unwanted or dopant atoms. We performed molecular dynamics simulations using machine learning Gaussian Approximation Potentials [3] trained on a large dataset of atomic structures generated with *ab initio* methods [4,5]. Our results show that incorporation of dopant atoms causes a significant change in the structure of aBN, which is strongly reflected in the thermal and mechanical properties of the compounds [4]. Experimentally, aBN films were synthesized by atomic layer deposition on silicon substrates, and their thermal conductivity was studied using a frequency-domain thermoreflectance setup.

Further, in the context of ultralow dielectric constant and interconnect technologies [1,2], it is crucial to understand how this structural complexity of aBN influences its electronic properties, which are also a first step towards the elucidation of its optical response and spectroscopic characterization. In this spirit, we discuss some early theoretical investigations towards this goal.

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Coupling Spin Defects in a Layered Material to Nanoscale Plasmonic Cavities

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Spin defects in hexagonal boron nitride, and specifically the negatively charged boron vacancy (V_B^-) centers, are emerging candidates for quantum sensing. However, the V_B^- defects suffer from low quantum efficiency and, as a result, exhibit weak photoluminescence. In this work, a scalable approach is demonstrated to dramatically enhance the V_B^- emission by coupling to a plasmonic gap cavity. The plasmonic cavity is composed of a flat gold surface and a silver cube, with few-layer hBN flakes positioned in between. Employing these plasmonic cavities, two orders of magnitude are extracted in photoluminescence enhancement associated with a corresponding twofold enhancement in optically detected magnetic resonance contrast. The work will be pivotal to progress in quantum sensing employing 2D materials, and in realization of nanophotonic devices with spin defects in hexagonal boron nitride.

Observation of 2D ferroelectric domains in folded hBN flakes

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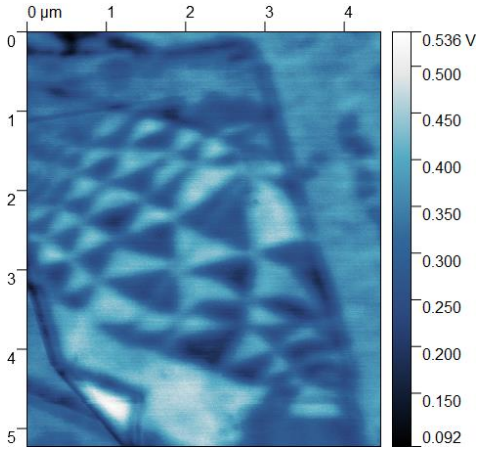
The degree of freedom offered by the control of the stacking angle between layers of 2D materials opened the way to a large variety of intriguing physics phenomena related to the so-called moiré potential or to surface reconstruction. Particular stacking arrangement enables to engineer physical properties that are totally absent in the parent material. One of the most famous study is the demonstration of superconductivity in twisted bilayer graphene [1]. For hBN, it has been shown that parallel stacking can give rise to 2D ferroelectricity [2–4]; an important building block for the integration of complex functionalities in van der Waals heterostructures. Indeed, when two hBN flakes are stacked with a twist angle close to 0°, spontaneous out-of-plane electric polarization occurs on large reconstructed triangular domains with AB and BA arrangements [5]. Nevertheless, fabrication of such artificial stacks usually relies on relatively complex transfer techniques. We will show that large ferroelectric domains can be observed on thin folded hBN flakes using Kelvin Force Microscopy (KFM) (see Figure). Integration of such 2D ferroelectric layers into more complex van der Waals heterostructures will be discussed.



Surface potential of a hBN flake probed by Kelvin Force Microscopy (KFM). Triangular ferroelectric domains with out-of-plane polarization corresponding to AB and BA stacking are visible on the folded part of the flake.

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Strain tuned non-classical light emission from localized defect states in 2D layered semiconductors

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Among various available non-classical light sources, localized defect states in two-dimensional (2D) van der Waals (vdW) materials, including semiconducting transition metal dichalcogenides, GaSe and insulating h-BN, have emerged as a promising physical system for scalable quantum technologies. However, in spite of significant advances in the field, the exact nature of these emitters is still under debate. In this work, the direct visualization of individual atomic-scale defects in h-BN flakes using atomic force microscopy under ambient conditions combined with density functional theory calculations of their band structures and electronic properties made it possible to associate the existence of several single-photon optical transitions to the observed defects, thus shedding light on the origin of quantum emitters in h-BN. Furthermore, we report on the dynamic real-time spectral tuning over a few-meV-wide range of the optical emission from individual non-classical light sources in h-BN flakes subjected to the radio frequency surface acoustic waves. Our approach provides an effective post-fabrication in situ tuning method capable of controlling otherwise random emission energy of 2D light sources that severely limits their suitability for any practical future applications in quantum photonics. We also demonstrate a scalable and lithography-free approach toward creating large areas of localized emitters in 2D semiconductors. The proof-of-concept was achieved by placing WSe_2 and GaSe flakes over polystyrene or luminescent rare-earth ion doped micro/nano-particles. Altogether, this study opens the door to the use of static and dynamic strain engineering for scalable integration of vdW emitters in nanophotonic and related quantum information technologies.

Characterization and Manipulation of Interfacial-hBN Emitters

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Point defects in the crystal lattice of 2D materials can produce optical, electrical, and quantum properties that significantly differ from those of the bulk crystal. Hexagonal boron nitride (hBN) is generally transparent but defects in the lattice structure create intra-band gap energy levels and result in localized emission of visible light, known as colour centres. In the interaction between the surface of hBN and an interfacial liquid layer there is a successive activation of quantum emitters, particularly in organic solvents. Emitters even seem to hop from defect to defect. STM has shown that these defects exhibit different charge states, and a comprehensive review of different liquids shows that the activation is largely dependent on the interacting liquid. The questions of how to probe and how to influence these surface interactions remain. Using spectral single molecule localization microscopy (sSMLM) we can localize these emitters with resolution in 10s of nanometres and determine emitter density, intensity, dynamics, and spectra with a wide-field microscope. To investigate the interfacial dynamics of these charged sites in situ, we employ an electrochemical cell adapted to our widefield spectral SMLM microscope, applying an out-of-plane electrochemical bias. Combining experimental manipulation with theoretical modelling can help elucidate these defects properties and realize extreme-confinement applications.

Van der Waals Heterostructure of Hexagonal Boron Nitride with an AlGaN/GaN Epitaxial Wafer for High-Performance Radio-frequency Applications

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While two-dimensional (2D) hexagonal boron nitride (h-BN) is emerging as an atomically-thin and dangling-bond-free insulating layer for next-generation electronics and optoelectronics, its practical implementation into miniaturized integrated circuits has been significantly limited due to difficulties in large-scale growth directly on epitaxial semiconductor wafer. Herein, the realization of wafer-scale h-BN van der Waals heterostructure with 2-inch AlGaN/GaN high-electron mobility transistor (HEMT) wafer by using Metal-Organic Chemical Vapor Deposition (MOCVD) is presented. The combination of state-of-the-art microscopic and spectroscopic analyses and theoretical calculations reveals that the hetero-interface between ~2.5 nm-thick h-BN and AlGaN layer is atomically sharp and exhibits a very weak van der Waals interaction without formation of a ternary or quaternary alloy that can induce undesired degradation of device performance. The fabricated AlGaN/GaN HEMT with h-BN shows very promising performance including the cut-off frequency (f_T) and maximum oscillation frequency (f_{MAX}) as high as 28 and 88 GHz, respectively, enabled by an effective passivation of surface defects on the HEMT wafer to deliver accurate information with minimized power loss. These findings pave the way for practical implementation of 2D materials integrated with conventional microelectronic devices, and the realization of future all-2D-electronics.

Liquid-activated quantum emission from native hBN defects for nanofluidic sensing

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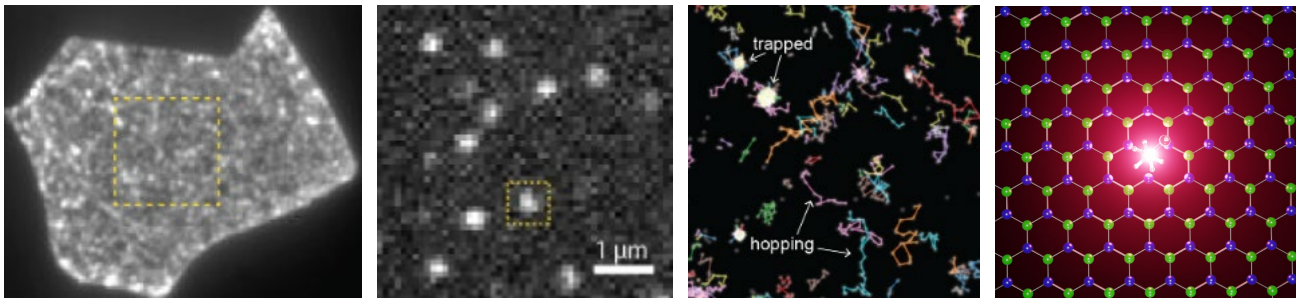
We report visible-range fluorescent emission from pristine hexagonal boron nitride (hBN) crystals immersed in organic solvents. We show that the emission comes from transient, liquid-activated quantum emitters, arising from the reversible binding of liquid molecules onto native defects present at the surface of hBN. These defects can be utilized as molecular sensors in liquid, without compromising the atomic smoothness of their host material. Vibrational spectra and the photodynamics of the emitters suggest that their activation occurs through the chemisorption of carbon-bearing liquid molecules onto native hBN defects. The correlated activation of neighbouring defects reveals single-molecule dynamics at the interface, while defect emission spectra offer a direct readout of the local dielectric properties of the liquid medium. We harvest these effects in atomically smooth slit-shaped van der Waals channels, revealing molecular dynamics and suggesting an increasing dielectric order under nanometre-scale confinement.



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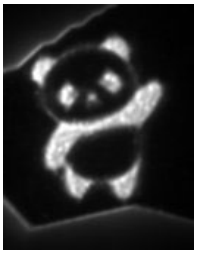

The pursuit of deep-UV defect emitters in 2D hBN

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Room temperature defect quantum emitters in hexagonal boron nitride have emerged as the source of considerable scientific interest. Recent studies have demonstrated the ability to engineer reliable single photon sources in thin film hBN with reproducible emission properties in all spectral ranges [1], suitable for applications such as quantum communications.
While such quantum emitters have been extensively studied for the visible and near-IR regime, this work aims to investigate the recently observed deep-UV luminescence of hBN point defects at 4.1eV, likely caused by carbon substitutions at nitrogen sites [2].
A frequency-quadrupled titanium-sapphire laser at 200nm is utilized for pulsed, above band-edge excitation, while the recombination dynamics of the UV defect emission are studied by temperature-dependent, time-resolved micro-photoluminescence spectroscopy. A direct dependence between the luminescence of the defect and its environment is explored to establish a relationship between the properties of the 2D material samples and the electron-phonon coupling. These findings are corroborated by Raman measurements.
Consequently, we are able to establish a scientific basis for further investigation into the UV emission of hBN with the prospect of identifying single photon emitters under optical excitation.

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Symmetric carbon tetramers forming chemically stable spin qubits in hBN

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Point defect quantum bits in semiconductors have the potential to revolutionize sensing at atomic scales. Currently, vacancy related defects, such as the NV center in diamond and the VB^- in hexagonal boron nitride (hBN), are at the forefront of high spatial resolution and low dimensional sensing. On the other hand, vacancies' reactive nature and instability at the surface raise concerns. Here, we study the symmetric carbon tetramers in hBN and propose them as a chemically stable spin qubit for advanced sensing in low dimensions. We utilize periodic-DFT and quantum chemistry approaches to reliably and accurately predict the electronic, optical, and spin properties of the studied defect. We show that the defects give rise to spin state dependent optical signals with strain sensitive inter system crossings rates. Furthermore, the relatively weak hyperfine coupling of the defect to their spin environments results in a reduced electron spin resonance linewidth that may enhance sensitivity. We show that the boron centered tetrameter can implement a spin qubit with highly desirable specifications in hBN.

Reduction of MOVPE h-BN/sapphire interaction by wrinkle formation revealed by Raman studies

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Van der Waals heterostructures are currently a hot topic due to their flexibility and the possibility of easy integration. In the case of such devices, phenomena occurring at material interfaces are of key importance. One important mechanism is the generation of strain between different materials due to differences in the lattice thermal expansion. The properties of such thin, strained layers are determined by the competition between the elastic energy of the material and the energy of the interaction with the substrate.

We discuss the interaction between MOVPE h-BN [1] and the sapphire substrate based on temperature-dependent Raman studies of the h-BN in-plane E_{2g}^{high} phonon mode. In literature, luminescence is known to hinder Raman measurements of MOVPE h-BN above 500 K [2]. However, we succeed in extending our research up to 1100 K since we found that this luminescence can be quenched by annealing samples. The analysis of the rate of the change of the E_{2g}^{high} phonon energy along with temperature indicates that below 900 K epitaxial h-BN interacts with sapphire 1-2 orders of magnitude weaker than at higher temperatures (Figure 1.). We attribute this drop to the creation of h-BN wrinkles. When the temperature is decreasing after the growth, differences in lattice thermal expansion of h-BN and the sapphire substrate lead to the generation of strain. Close to 900 K, the critical strain results in wrinkles formation. Below this temperature, almost no additional strain is generated but new wrinkles are created [3]. The presence of wrinkles is an indicator of van der Waals epitaxy resulting from a weak interaction with the substrate and is inherent to high-temperature growth. An understanding of the process of wrinkle formation is therefore crucial for any future growth of van der Waals heterostructures.

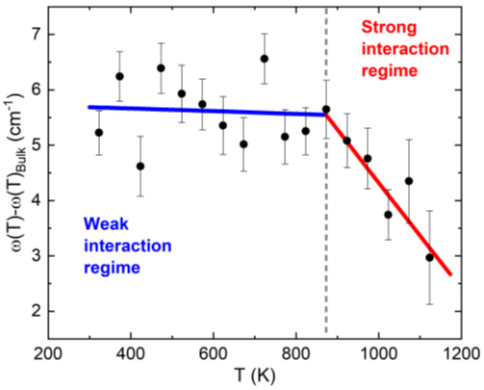

Figure 1. Differences between in-plane phonon energies in as-grown MOVPE h-BN and bulk material. Black dots – experimental points. Blue (red) curve – fitted theoretical formula in the regime of weak (strong) h-BN/sapphire interaction.

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Surface recombinations and out of plane diffusivity of free excitons in hexagonal boron nitride

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Using cathodoluminescence (CL) experiments as a function of the beam incident energy, we measured the out-of-plane diffusion length (L) of free excitons in bulk hBN and the recombination properties of its surface. The exciton lifetime (τ) is measured independently with a home-made time-resolved CL setup at GEMaC [1], and the diffusion constant (D) is extracted from the relation $L = \sqrt{D\tau}$. The diffusion properties are analysed on samples from different synthesis processes (High Pressure High Temperature (HPHT) [2], Atmospheric Pressure High Temperature (APHT) [3], Polymer Derived Ceramisation (PDC) [4]) and at different temperatures. The exciton diffusivity appears limited by scattering on impurities or defects.

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Study of the effects of nanoscale environments on single emitter's fluorescence properties in hexagonal Boron Nitride

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Hexagonal Boron Nitride (hBN) is often discussed as a promising platform for quantum information technologies. One of the obstacles lies in the heterogeneity and stability of the singe photon emitters (SPE), which is still challenging as recent studies find [1]. We aim to understand the effect of surface interactions, controlling the nanoscale environment of SPEs by exposing hBN to various liquids and using a combination of hyperspectral confocal microscopy and widefield TIRF microscopy to analyze the resulting effects on the fluorescence properties. The hBN samples were prepared by a scalable atmosphere pressure CVD fabrication approach, that was shown to produce high quality hBN [2]. The hyperspectral analysis shows that our material harbors the hBN characteristic emission spectra and heterogeneity.

To analyze the widefield TIRF image series recorded with 50 ms per frame we use super-resolution microscopy algorithms to extract SPE in hBN-associated time traces, recorded in air and after addition of selected liquids, namely water, IPA, ethanol, and hexane.

We analyse surface density, mean intensity, and power law blinking for hundreds of SPEs in dependence of the excitation wavelengths (488, 561 to 647 nm). The results indicate that immersing hBN in any of the liquids activates large numbers of SPEs. Interestingly the SPE intensity may increase or decrease in dependence of the specific solvent and excitation wavelength used, while nonpolar solvent hexane was always associated to an intensity decrease. This work proposes an avenue in providing a detailed understanding of SPE properties and control required for integration in quantum devices.

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Quantum sensing with spin defects hosted in a van der Waals material

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Quantum sensing technologies based on solid-state spin defects have already shown a huge potential to cover the growing need for high-precision sensors [1], both for fundamental research and for industrial applications. The most advanced quantum sensing platforms to date rely on optically- active spin defects embedded in three-dimensional (3D) materials. A prime example is the nitrogen- vacancy (NV) center in diamond, which has already found a wide range of applications in condensed matter physics, life sciences and geophysics [2]. Despite such success, NV-based quantum sensing technologies still face several limitations that mainly result from the 3D structure of the diamond host matrix. They include (i) a limited proximity between the quantum sensor and the target sample, which hampers its sensitivity, and (ii) the inability to engineer ultrathin and flexible diamond layers, precluding an easy transfer of the quantum sensing unit onto the samples to be probed as well as its integration into complex multifunctional devices. An emerging strategy to circumvent these limitations consists in using spin defects embedded in a van der Waals crystal that could be exfoliated down to the monolayer limit. Such a 2D quantum sensing foil would offer atomic-scale proximity to the probed sample together with an increased versatility and flexibility for device integration.

Hexagonal boron nitride (hBN) is currently the most promising van der Waals crystal for the design of quantum sensing foils [3]. This insulating material, which can be easily exfoliated down to few atomic layers while maintaining chemical stability, is extensively used for encapsulation of van der Waals heterostructures. Furthermore, hBN hosts a broad diversity of optically-active point defects owing to its wide bandgap [4]. In this work, we focus on the negatively-charged boron-vacancy (VB^-) center in hBN, which features magneto-optical properties very similar to those of the NV defect in diamond, with a spin triplet ground state whose electron spin resonance (ESR) frequencies can be measured via optically-detected magnetic resonance methods at room temperature [5, 6]. We analyze the performances of thin hBN flakes doped with VB centers for quantitative magnetic field imaging in van der Waals heterostructures [7]. As a proof-of-concept, we image the magnetic field produced by CrTe2, a van der Waals ferromagnet with a Curie temperature above 300 K. We then investigate how the properties of VB centers evolve with the hBN thickness down to the monolayer limit.

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Creation and photophysical analysis of blue single photon emitters in hexagonal Boron Nitride

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Abstract

Hexagonal Boron Nitride (hBN) is a III-V two-dimensional van der Waal insulator material that is known to host single photon emitting colour centers in the visible range. Although these defects are stable, bright and exhibit high purity, they lack from variation in spectral repdocuceability, spectral diffusion and deterministic fabrication. Recent studies reported creation of blue spectral range single photon emitters in hBN using electron beam irradiation and cathodoluminescence characterisation [1]. These defects, known as blue emitters, display consistent zero phonon transition at 436 nm, low spectral diffusion [2] and insusceptibility to electric fields [3]. However, conducting photophysical properties of blue emitters is necessary to understand their energy levels transitions and gain insight into presence of any metastable states.

The detailed photophysical analysis of the blue emitters in hBN covered aspects such as photostability, saturation behavior, lifetime, and transition rates, and was performed using time-resolved excitations and time correlation measurements. The measured blue emitters were created by electron beam irradiation in three different hBN flakes, which were annealed under different conditions. Through photon statistics and photodynamic analysis, possible level structure of the emitter was revealed, suggesting that a lack of a metastable state, which is supported by theoretical analysis proposing that the defect may have an electronic structure featuring a fully occupied defect state in the lower position of the bandgap and an empty defect state in the upper half of the bandgap.

The majority of the emitters showed outstanding photostability over four hours and behaved like an ideal two-level system. The zero-phonon lines (ZPLs) of emitters is consistently 436 nm and the lifetime of the blue SPEs is approximately 2 ns. In summary, our findings have significant implications for comprehending the photophysical properties of the new class of blue quantum emitters in hBN, which have the potential to act as a promising source for quantum applications.

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Epitaxial growth of hexagonal boron nitride on silicon carbide and sapphire by high-temperature molecular beam epitaxy

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According to the empirical rule about the relation between the optimal growth temperature T_G and the melting point T_M of material, $T_G \sim T_M/2$, hBN growth temperature is suggested to be around 1500 °C.[1] On the other hand, it is often said that 2D materials such as hBN has less restrictions on substrate choice by virtue of weak van der Waals interaction with the substrate. In this study, we compare two BN samples grown at a high T_G (> 1400 °C) on 4H-SiC substrates under nominally identical conditions, but except for the presence and absence of epitaxial graphene on the substrate. Despite such a high growth temperature, there were clear distinctions between the two samples: only the sample grown on epitaxial graphene showed a clear layered structure with the sp^2 bonding character, while the BN directly grown on SiC without epitaxial graphene showed poor crystallinity, implying that the presence of such 2D substrate template will be critical for layered material overgrowth. In contrast, we have recently demonstrated that clear layered hBN can be grown on c-plane sapphire substrate.[2] The successful growth of such sp^2-bonded hBN on sapphire substrate with no apparent 2D layer now becomes puzzling from the point of view of the aforementioned BN grown on SiC. Interestingly, however, we should mention that when BN growth was initiated on the sapphire unintentional AlN nucleation layer was spontaneously formed before the intended hBN layer growth.[2] We will discuss the role of such ultra-thin AlN on the hBN overgrowth and a possible strategy for large-area hBN epitaxy.

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Fabrication of spin defects in hexagonal boron nitride by focused ion beams

hBN is fast becoming the material of interest as a host for optical active defects owing to its wide bandgap and two dimensional nature. Most studies of the spin-photon interface in hBN focus on the negatively charged boron vacancy (V_B^-) defect, which is typically fabricated by ion irradiation. V_B^- fabrication methods however lack robustness and reproducibility when applied to thin flakes ($\lesssim$10 nm) of hBN. Thin flakes are crucial for high sensitivity applications, as the defects can be close to both the subject and the collection path. Here we identify mechanisms that both promote and inhibit V_B^- generation and optimize ion beam parameters for site-specific fabrication of optically active V_B^- centers. We used a novel approach towards fabrication, via irradiation with a proton beam, and placing the sample on an angle to achieve surface irradiation. Previous methods with normal incidence nitrogen beams have been too destructive, destroying the hBN lattice beyond repair. The impact of the substrate on the defect generation is also investigated, comparing the intensities of the V_B^- defect in suspended and supported flakes. We accentuate conditions accessible by high resolution focussed ion beam (FIB) systems, and present a framework for V_B^- fabrication in hBN flakes of arbitrary thickness for applications in quantum sensing and quantum information processing.

Quantum Key Distribution Using a Room Temperature Integrated Single Photon Source in Hexagonal Boron Nitride

Hexagonal Boron Nitride (hBN) is one of the various solid-state materials that is capable of hosting atomic defects which can emit high-purity single-photons. Reliable, deterministically generated single photons on demand are a prime candidate for quantum-based technologies such as quantum key distribution (QKD). QKD enables the sharing of secret keys between distant users and utilises photons as information carriers. The current challenge is to implement these protocols in practice for real work conditions in a compact manner. In this study, we integrate single photon sources (SPS) in hBN with solid immersion lenses (SILs) and explore a robust, scalable way to increase the signal to noise ratio and combat the problem of collection efficiency, the room temperature SPS is used with a discrete-variable quantum key distribution system operating in free space. We demonstrate employing the source in the "plug and play" system, under realistic conditions by generating keys with one million bits length and demonstrated secret key of approximately 70,000 bits and a quantum bit error rate of 6% and ε-security of 10^{-10}. With an emphasis on including all known effects impacting derived sercurity level, demonstrating the most trustworthy QKD system realised with SPSs to date. We also show that high secured QKD rates approaching MHz are tangible over several kilometres with quantum bit error rate simulations.

Insight into the nature of blue emitters in hexagonal Boron Nitride via Stark effect

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Single photon sources 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 quantum emitters in the visible wavelength range, but it has only recently become possible to site-specifically generate blue quantum emitters within hBN lattice[1]. These blue emitters have a uniform wavelength of 436 nm and exhibit GHz-range linewidths, due to effects such as spectral diffusion. Here, we perform coherent photoluminescence excitation to measure the Stark shift under an in-plane or out-of-plane electric field, and map out the electric dipole interaction to determine the likely defect structure.

The Stark effect generated from an applied electric field on a quantum emitter is estimated from the Lorentz local field approximation given by $\Delta E = -\Delta\mu F - \frac{1}{2}\Delta\alpha F^2$ (1), where F is the electric field (MV/m), and $\Delta\mu$ and $\Delta\alpha$ are changes in permanent dipole moment and in polarizability, respectively, of the ground and excited states. Vertical and horizontal devices were fabricated to apply out-of-plane and in-plane electric fields, respectively. Both devices were cooled to 5 K, and considering the narrow inhomogeneous linewidth of blue emitters, measurements were conducted on resonance with high spectral resolution. From our results we observed an insignificant linear shift in the out-of-plane electric field orientation and a large quadratic response in the in-plane configuration. According to Equation (1), it was deduced that the observed wavelength shift of blue emitters has minimal contribution from permanent transition dipole moments, but is mainly governed by the second term, transition polarizability. In comparison to other studied quantum emitters in hBN, which emit at longer wavelengths[2,3], our findings reveal that blue emitters are less susceptible to local field variations in the lattice due to small permanent dipoles. The study concluded with a proposition of likely candidates for blue emitter defects via density functional theory (DFT) modelling.

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Impact of bubble creation on optical properties of h-BN

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Similarly to other 2D materials, h-BN can be easily deformed e.g. by applying strain or by transferring it on a pre-patterned substrate. It has been shown that a strong bending of h-BN can activate the quantum emission [1] due to the presence of a trapping deformation potential and can also influence other optical properties. An interesting way of h-BN deformation is to create bubbles. Since they exhibit non-uniform strain distribution [2], their optical properties are also inhomogeneous. Therefore they bear great hope for both the activation and tuning of the properties of quantum emitters.

We discuss the optical properties of bubbles created via electron irradiation of our MOVPE h-BN grown on sapphire substrates [3,4]. Photoluminescence (PL) measurements are performed using different excitation wavelengths to study the influence of a deformation induced by the presence of bubbles on defect-related emission in various spectral ranges. PL mapping measurements (Fig. 1) reveal a strong enhancement of light emission on bubbles as well as a shift of the E_{2g} phonon mode energy (which is an indicator of strain). It is found that creating bubbles not only induces a strong increase in the total PL intensity but also selectively enhances some spectral lines which are difficult to observe for flat, non-deformed material. Combining the above with the possibility of creating h-BN bubbles in a precisely selected location on demand, we believe h-BN bubbles can become a useful tool to deterministically locate and activate quantum emitters.

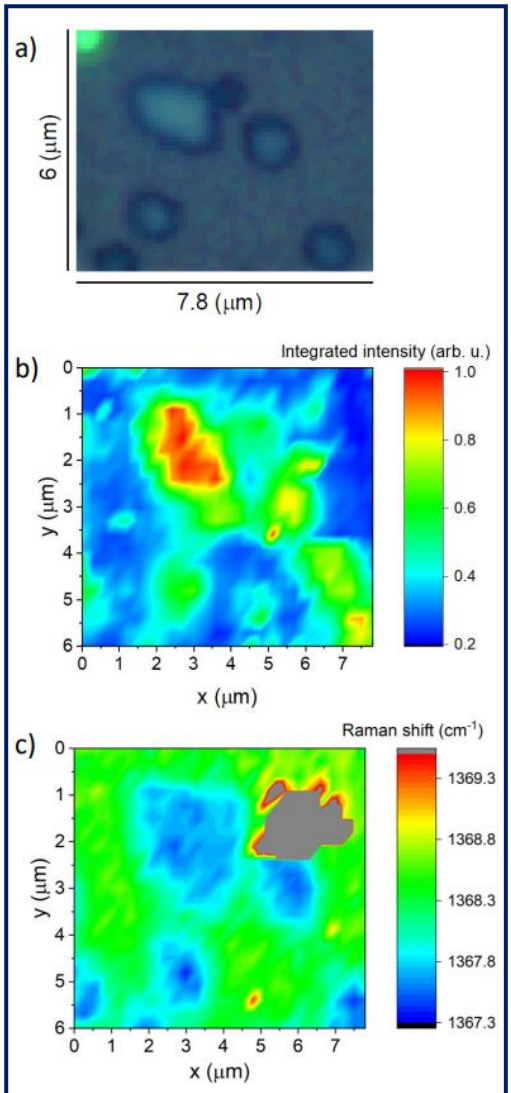

Fig. 1. Room temperature PL measurements of h-BN bubbles with 532 nm laser excitation: a) optical image, b) total integrated intensity, c) Raman shift of the in-plane phonon mode

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Electronic Structure of h-BN under Stacking, Folding, and Twisting Deformations

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The optical properties of two-dimensional (2D) semiconductors can be easily influenced by low energy deformations, such as layer gliding, twisting, bending, or folding. In folded h-BN layers, two types of structural modifications occur: local curvature at the folded edges and interlayer shear of the layers that changes the stacking of the overlapping flat regions. In this work, we demonstrate that the overall electronic behavior of folded h-BN layers can be described as a combination of these two effects, which both induce local bandgap changes. Specifically, an h-BN folded monolayer can be viewed as a type-II junction between two wide gap semiconductors located at the curved and flat overlapping zones [1].

Although changes in the h-BN stacking induced by a basal plane glide can modify the exciton response [2], these structures are rarely observed in transmission electron microscopy. Instead, twisted layers are more commonly encountered. Here, we unfold the folded and densely packed electronic bands of moiré h-BN supercells over the primitive Brillouin zone, which allows us to reveal perturbations induced by the twist of the layers on the pristine perfect system band structure. Finally, we employ many-body perturbation theory techniques (GW+BSE) to show the effect of twisting on the excitonic response of h-BN bilayers.

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Probing intrinsic properties of epitaxial monolayers of h-BN on graphite with scanning tunnelling microscopy

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Hexagonal boron-nitride (h-BN) has gathered great scientific interest over the past years, as its wide electronic band gap and single-photon emission (SPE) at room temperature offer great opportunities for developing novel applications[1]. Regardless of these recent efforts towards the characterization of h-BN, intrinsic aspects of this layered material remained elusive, such as the magnitude of its electronic band-gap and excitonic binding energy. Additionally, the crystallographic nature and impacts of the point defects responsible for the SPE are still under discussion[2]. In this study, we used scanning tunnelling microscopy/spectroscopy (STM/STS) combined with optical spectroscopies to characterize monolayers of h-BN epitaxially grown on graphite by high-temperature molecular beam epitaxy[3]. On defect-free regions, these measurements revealed an electronic band-gap of (6.8 ± 0.2) eV, which, in combination with deep UV optical spectroscopy leads to a (0.7 ± 0.2) eV exciton binding energy. Furthermore, STM imaging revealed the presence of point defects, whose electronic and optical properties were probed by a combination of STS, photoluminescence and STM-Cathodoluminescence measurements. We understand that this approach paves the way for the long-sought goal of identifying the nature of the defects responsible for the SPE on h-BN.

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Optical characteristics of single-defect colour centres in hexagonal boron nitride

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Colour centres in two-dimensional hexagonal boron nitride (hBN) are a promising candidate towards a 2D spin-photon interface, offering the advantage of room temperature single-photon emission combined with integration into scalable and compact hardware, thanks to their reduced dimensionality.

Realizing a spin-photon interface in this material relies on identifying a defect species with highly coherent spin and optical degrees of freedom. Although both room-temperature access to single-defect spins and room-temperature Fourier-transform limited optical emission have been observed for spin defects in hBN, it is unclear if the defects in these separate investigations are related in structure.

We present work on a spin-active carbon-related defect species showing promising optical properties at room temperature, with the goal of determining the limits of single-defect optical coherence. Via temperature-dependent spectroscopy of individual defects, we investigate the mechanisms responsible for broadening of the single-defect optical emission from room temperature down to cryogenic conditions.

These results are an important step towards realizing coherent excitation of these defect centres. Additionally, understanding the photo-dynamics of these emitters provides additional information towards the identification of their microscopic structure, and will help unravel their potential as solid-state qubits and nanoscale quantum sensors.

Spin physics of single defects in hexagonal boron nitride

Simone Eizagirre Barker[1*], Hannah Stern[1], Oliver Powell[1], Carmem M Gilardoni[1], Xiaoxi Deng[1], Qiushi Gu[1], Louis Follet[1], Chi Li[2], H H Tan[3], Igor Aharonovich[2], Mete Atatüre[1]

Single-photon emitting defects in hexagonal boron nitride (hBN) have emerged as a promising platform for quantum photonic applications, as they can be readily integrated into nanoscale devices and sensors thanks to their reduced dimensionality and show promising optical and spin properties at room temperature.

Recently, we showed the first demonstration of optically-detected magnetic resonance (ODMR) from single spin defects in a van der Waals material at room temperature, with ODMR contrasts of up to 30%, demonstrating their potential as solid-state qubits. In this contribution, we present our latest results towards determining the spin model of these singly addressable defects and further reveal their potential for applications in quantum information and sensing.

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Electronic and optical properties of boron nitride in the wurtzite phase

Abstract

Wurtzite polymorph of boron nitride (wBN) was first obtained in the 1960s starting from hexagonal boron Nitride (hBN) under high pressure. Since then, several studies have been conducted on samples obtained mostly by use of the shock compression technique which allowed the production of micrometer BN powder of both wurtzite and cubic phases. Moreover, wBN has long been recognized as a metastable material since it tends to undergo a transition back to the hexagonal phase. Recently, it has been shown that it is possible to produce larger and purer samples of wBN and that they can be stabilised by macroscopic defects in the crystals. Since this material is a wide-band-gap semiconductor believed to have a hardness comparable to that of cubic boron nitride (cBN) and diamond, its implementation in optoelectronic devices in harsh environments can be envisaged. We characterized the electronic properties of wBN by use of the quasi-particle formalism in G_0W_0 approximation on top of Density Functional Theory in Generalized Gradient Approximation (DFT+GGA) in order to take into account screening effects usually neglected in standard DFT calculations. Optical properties were studied by mean of the Bethe-Salpeter equation (BSE). Finally, we studied the Boron vacancy in wBN since its wide band gap makes it a good candidate to host defect levels that can find applications in quantum technologies.

Decoherence and multi-spin dynamics of the VB^- center in hBN

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Unlike any other point defect quantum bits, the negatively charged boron vacancy (VB^-) center in hexagonal boron nitride (hBN) interacts with many close nuclear spins. The relatively strong coupling and the vast phase space of the spin system have both advantageous and disadvantageous consequences on the dynamics of this center. In my contribution, I discuss the decoherence and the theory of multi-spin dynamics of freely evolving and microwave driven VB^- spins. Relying on cluster correlation expansion calculations, I explore the limits of the Hahn echo coherence time at various nuclear spin abundances and magnetic field regimes.[1] Furthermore, I discuss the case of microwave driven VB^- centers and analyze the contribution of the closest nitrogen nuclear spins to the dynamics of the system.[2] Finally, I present our latest results on dynamic nuclear polarization enabling hyperpolarization of distinct nuclear spins.[3]

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Tuning of hBN bandgap by aluminum alloying

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In our research we focus on filling the efficiency gap in the DUV spectral range. We test an approach based on hexagonal boron nitride which despite of an indirect bandgap has a photoluminescence (PL) intensity of the band-edge transition that is more than two orders of magnitude higher than that of an AlN epilayer which has a direct bandgap [1]. Because the conduction band minima responsible for direct and indirect emission are close to each other in hBN, the emission efficiency can be potentially further enhanced when electronic states are properly manipulated [2].

In this work we study boron nitride layers grown with metalorganic vapor phase epitaxy (MOVPE) on 2-inch sapphire substrates [3-4]. The studied samples were alloyed with aluminum using different flows of trimethylaluminium (TMAl). Based on results obtained by UV-Vis, Raman and Fourier-transform spectroscopies, X-ray diffraction, scanning electron microscopy, atomic force microscopy and photoluminescence we will discuss how different flows of aluminum precursor affect the properties of the final epitaxial hBN layer.

As presented in the figure 1, the absorbance spectra changes with the amount of TMAl. The spectra revealed two peaks which energies coincide with direct (higher energy) and indirect (lower energy) transitions in hBN. The intensity ratio and the position of those peaks vary between the samples. This observation suggests that we are capable of manipulating the boron nitride bandgap by alloying with aluminum. The possible reasons and consequences of such a behavior will be discussed.

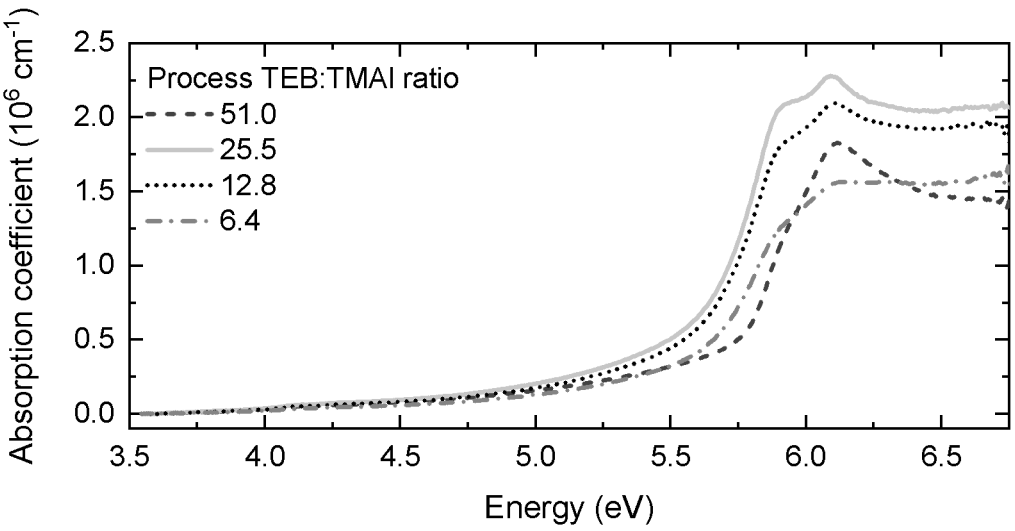

Figure 1: Absorption coefficient measured at room temperature for the samples grown with different TEB:TMAl ratio. Absorption coefficient uncertainty is estimated to be 10^5 cm^{-1}.

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Impact of oxygen on hBN nanowalls synthesis

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hBN can be synthesized into nanostructured films such as nanowalls by the introduction of hydrogen into plasma of physical vapor deposition (PVD)[1]. The oxygen-assisted hBN (O-hBN) nanosheets exhibit a high adsorption rate and capacity for metallic ions due to the unique polarity of B-O bonds and boron vacancies, surpassing the bulk and activated BN absorbents[2]. In this work, the influence of oxygen on hBN nanowall structural and optical properties were investigated. Oxygen gas in concentrations ranging from 0% to 0.1% was added to the PVD plasma of a RF sputtering system along with an $Ar/N_2/H_2$ gas mixture wherein-, O-hBN films were deposited at 95°C on Si (100) substrates. The surface morphology of O-hBN films changes from nanowalls to a granular structure at 0.1% O_2. Due to the higher affinity for oxygen, boron atoms bond easily with oxygen atoms forming more B-O bond forms at the expense of B-N bonds, as evident from the Fourier transform infrared spectroscopy. The characteristic Raman spectroscopy peak intensity of hBN at 1368 cm^{-1}, attributed to E_{2g} phonon vibration mode, decreases with increasing oxygen gas percentage during film deposition, thereby indicating a decrease in the crystalline nature of the hBN phase in O-hBN films. The O-hBN films become more hydrophilic as determined via the sessile drop contact angle method, with increasing O_2 percentage during the deposition. The OhBN nanowalls with high surface area and hydrophilic properties could potentially adsorb and remove toxic metallic ions like Pb^{2+} than other adsorbents like activated carbon for water purification applications.

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We report the characterisation of hBN thin films grown on sapphire substrates using high-temperature molecular beam epitaxy (HT-MBE). We have grown hBN films both with, and without, intentional *in-situ* carbon doping and measured them using variable angle spectroscopic ellipsometry (VASE), x-ray photo emission spectroscopy (XPS), atomic force microscopy (AFM) and confocal fluorescence spectroscopy (CFS). The optical constants of the undoped HT-MBE grown hBN films determined by VASE are similar to those measured previously by imaging ellipsometry on flakes exfoliated from hBN bulk crystals. There is a sharp onset of optical absorption close to 6 eV demonstrating the high quality of the undoped films. When measuring hBN films with thicknesses below 5 nm it is difficult to obtain both the thickness and optical constants of the films using VASE, therefore the thickness of these films has been determined using XPS and AFM. At low levels of carbon doping, CFS and Hanbury-Twiss-Brown measurements identify single photon emitters in the hBN films with spectroscopic characteristics (570-590nm fluorescence when using 532nm laser excitation) similar to those observed at known carbon defects introduced into exfoliated hBN. At high carbon doping levels, hBN optical constants show evidence of optical absorption at photon energies around 4.3 eV and 5.4 eV. In these hBN films single photon emitters are no longer observed, and spatially homogeneous fluorescence is observed. The results indicate that the HT-MBE grown hBN films can be controllably doped with carbon to achieve hBN with single photon emitters.

Boron vacancy pair in hexagonal boron nitride: a novel quantum sensor

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Point defect qubits in layered semiconductors offer an attractive alternative to traditional approaches such as the nitrogen vacancy center in diamond, as their mature exfoliation processes can be utilized to host defects directly on the surface or in monolayer samples. Such advances make it possible to realize qubits with improved sensing accuracy and spatial resolution. In this contribution, we discuss the boron vacancy pair (VB2 center) in hexagonal boron nitride as a novel quantum sensor whose stability we confirmed not only by model calculations but by electron microscope experiments as well [1]. The low symmetry configuration of VB2 center results in unique electronic and spin properties, with the potential to implement an optically addressable spin qubit with unprecedented characteristics. The stress absorption capabilities of VB2 center can further tailor the qubit's electronic structure, thus enabling control over all of its magneto-optical properties. The possibility of long-term storage of population information in the singlet state, and the efficient decoherence protection applicable in the dense nuclear spin host make VB2 center a unique qubit candidate.

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REMOTE ELECTRON-PHONON AND PLASMON-PHONON INTERACTIONS IN BN-ENCAPSULATED GRAPHENE

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The widespread use of BN as an encapsulator raises the question of its interaction with the active layer. We focus here on remote electron-phonon interactions in van der Waals heterostructures, using density-functional theory (DFT). The phonons of BN capable of coupling remotely to electrons in other layers are identified, and the interaction is quantified as a function of the number of layers. The main challenge is the prohibitive cost of full DFT calculations on the whole heterostructure. To avoid it, the bare electron-phonon interactions originating from each layer is extracted from DFT [1] and then screened with the dielectric response from the heterostructure computed within a semi-analytical model [2]. BN's LO phonons couple remotely via the well-known Fröhlich interaction. BN's ZO phonon also couple remotely. In fact they only couple remotely, because the associated perturbation is odd with respect to the plane of the BN layer, implying a vanishing intralayer coupling. The current method, specifically tailored to study remote interactions via the behavior of the coupling potentials in the out-of-plane direction, is thus particularly useful in this case. The method is applied to BN/graphene heterostructures, accounting for the dynamic electronic response of graphene in the form of standard electronic screening as well as the coupling between graphene's plasmons and BN's phonons. While strongly screened most of the time, the coupling of graphene's electrons with BN's phonons may become much more significant when phonon momentum and energy fall around graphene's plasmon dispersion. Electron-phonon scattering is included in the Boltzmann transport equation to reveal the influence of this mechanism on transport.

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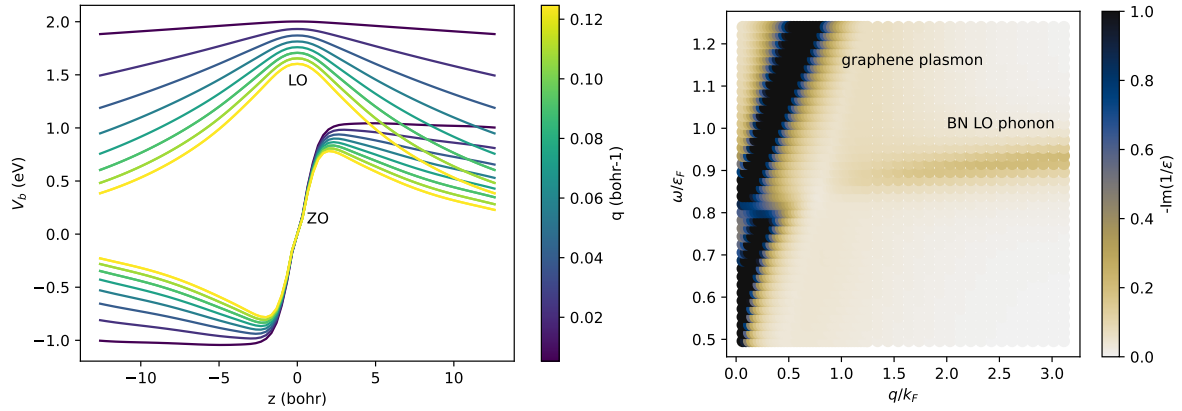

Figure 1: (Left) Perturbing potentials associated to the LO and ZO phonons of a single layer of BN in the x-y plane at $z=0$. (Right) Plasmon-LO phonon coupled modes in the graphene/5-BN heterostructure, revealed by the imaginary part of the inverse dielectric function.

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Wafer-scale growth of amorphous boron nitride thin film

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Amorphous materials have been widely studied in diverse research fields because of their disordered structures and properties different from crystalline materials. Among them, amorphous boron nitride (aBN) is attracting attention because of its excellent electrical (ultralow dielectric constant of 1.89, high breakdown field of 7.3 MV/cm) and mechanical properties (hardness > 11 GPa). However, since it is hard to synthesize aBN thin films with pure amorphous phase, there is difficulty in application for practical electronic devices. In this presentation, we show that 6-inch-scale aBN thin films can be grown by the plasma enhanced chemical vapor deposition (PECVD) and applied as a copper interconnect capping layer. aBN thin films are deposited at a back-end-of-line (BEOL) compatible process temperature (400 ℃). We found that the formation of hBN nano-crystallites in the aBN thin films results in the increase of dielectric constant as the thickness of the aBN thin films increases. Thus, we show the relationship of the thickness of the aBN thin film, the amount of hBN nano-crystallites, and the dielectric constant of the aBN thin film. In addition, aBN thin film blocks copper (Cu) diffusion at temperatures up to 500 ℃, indicating that aBN thin film can be used as a capping layer for Cu interconnects. aBN thin film also has high thermal stability up to 1,000 ℃. Our results can lead up to the practical application of aBN as an ultra-low dielectric material for electronic devices.

First-principles theory of quantum defects in hexagonal boron nitride

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Two-dimensional materials (TDMs) have been recently found to host a variety of quantum point defects that are potentially useful for advancing quantum information science and technology. Among a growing suite of the TDMs, hexagonal boron nitride (h-BN) has emerged as a host of bright single-photon emitters (SPEs) and optically active spin qubits operating at room temperature. However, there are several challenges to be addressed for realizing h-BN-based quantum applications. In this poster, we summarize our recent efforts to predict and understand optically active quantum defects in h-BN using first-principles density functional theory (DFT) [1-4]. In the first part of the poster, we discuss the property of SPEs in terms of their coupling behaviors to an electric field [1,4] and lattice strain [2] how these could be used to understand the structural and optical properties of SPEs. We then propose a new type of spin qubit candidates in h-BN [3]. We combine DFT and quantum embedding theories to show that out-of-plane X-Y dimer defects (X, Y=C, N, P, Si) form a new class of high-symmetry spin-triplet defects in h-BN. Our results broaden the range of quantum defect candidates in h-BN and strengthened the potential of h-BN quantum defects as materials platforms to realize TDM-based quantum sensors and integrated quantum photonics.

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Temperature-dependent energy-level shifts of spin defects in hexagonal boron nitride

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Abstract: Two-dimensional hexagonal boron nitride (hBN) has attracted much attention as a platform for realizing integrated nanophotonics, and collective effort has been focused on spin defect centers. Here, the temperature dependence of the optically detected magnetic resonance (ODMR) spectrum of negatively charged boron vacancy (V_{B^-}) ensembles in the range of 5-600 K is investigated. The microwave transition energy is found to decrease monotonically with increasing temperature and can be described by the Varshni empirical equation very well. Considering the proportional relation between energy-level shifts and the reciprocal lattice volume (V^{-1}), thermal expansion might be the dominant cause for energy-level shifts. We also demonstrate that the V_{B^-} defects are stable at up to 600 K. Moreover, we find that there are evident differences among different hBN nanopowders, which might originate from the local strain and distance of defects from the flake edges. Our results may provide insight into the spin properties of V_{B^-} and for the realization of miniaturized, integrated thermal sensors.

Keywords: van der Waals materials, point defect, electron spin resonance, Varshni empirical equation, thermal expansion, temperature sensor

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Growth of high-quality multilayered hexagonal boron nitride with the assistance of metal-B alloy

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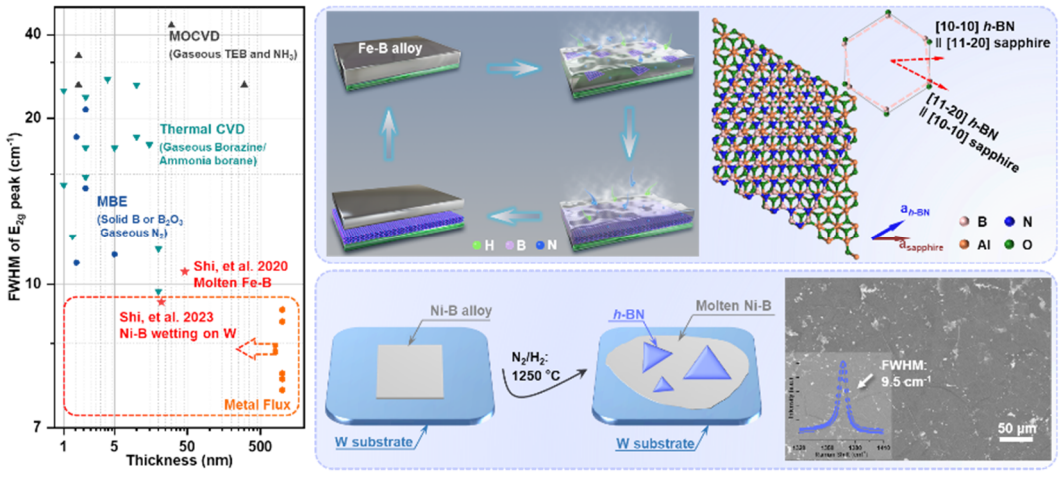
Fully exploiting the intrinsic characteristics and developing the applications of hexagonal boron nitride (*h*-BN) require an economically strategy to produce this significant material with high quality and low cost. With the assistance of metal-boron alloy, the controllable synthesis of large-area and uniform *h*-BN films has been recently achieved, representing important progress in an economic and environmentally friendly process. We reveal the vacancy-assisted growth mechanism of *h*-BN on Fe_2B substrate. It is found that B vacancies created by the formation of BN dimers play important roles in the migration of B and N atoms near the catalyst surface. We develop vapor-liquid-solid growth strategy to realize the epitaxial growth of multilayer *h*-BN on *c*-facet sapphire substrate using molten $Fe_{82}B_{18}$ alloy and N_2 as the source materials. We introduce molten Ni-B alloy wetting on W, the alloy serves as both the B source and the growth substrate for the synthesis of *h*-BN multilayers. The full width of half maximum of E_{2g} mode reaches 9.3 cm^{-1}, which is similar to the values of 8-9 cm^{-1} typically achieved by mechanically exfoliated bulk *h*-BN. The proposed metal-boron alloys yield multilayer *h*-BN with high quality and low cost, paving the way to potential applications in two-dimensional electronic and optoelectronic devices.



High-Q Polaritonic Resonators for Dielectric Sensing

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Polaritons allow for squeezing light at the nanoscale enhancing light-matter interactions, thus showing potentials for applications in spectroscopy or bio-sensing. Particularly, phonon polaritons (PhPs) in van der Waals materials, such as hexagonal boron nitride (h-BN), have attracted much interest because of their ultra-long lifetimes in the picosecond range and hyperbolic propagation with extremely large density of optical states [1-4]. Although propagating and dipolar-like PhPs modes have been studied in h-BN semi-infinite slabs and nanoresonators, respectively, high-order modes in h-BN resonators, with presumably much higher quality factors (Q) and field confinement, remain unexplored. Here, by infrared nano-imaging, we study, for the first time, the excitation and field distribution of high-order Fabry-Perot modes in h-BN resonators and realize in-situ sensing of the local dielectric environment. For the latter, we either transfer the same h-BN resonator on different polar substrates, such as SiO2 and SiC, or cover them with thin layers of another vdW material, such as high refractive-index transition metal dichalcogenides (e.g. WSe2). Our results provide insights into high order Fabry-Perot resonances in polaritonic structures as well as their functionality for in-situ local dielectric sensing, with applications in materials science and bio-sensing [5].

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CVD synthesis of sp^2-hybridized multilayer boron nitride films

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Since graphene isolation in 2004, the 2D materials is a blooming research field. Due to its unique properties, sp^2 hybridized boron nitride (BN) has been acknowledge as a key towards integration of other 2D materials in devices. Indeed, it is structurally very close to graphene – their lattice mismatch is only 1.7%- a semiconductor, atomically flat and thermally and chemically inert. It is therefore a choice material to be used in the van der Waals heterostructures with other 2D materials either as a top layer to protect another 2D material from its environment [1], or as a dielectric interlayer [2] and mostly, as a flat substrate [3]. However, these applications have been demonstrated using mechanically exfoliated BN from low defective and highly crystalline single crystals. Yet, this process limits the size of the devices that can be created to sub millimeter scale. In order to develop devices at a wafer scale, it is therefore critical to master the synthesis sp^2 hybridized BN layers at low cost, large scale and high quality.

In that respect, the goal of the researches we have undertaken is to develop the synthesis of sp^2-hybridized multilayer BN films with structural specifications fitting these requirements. We have already successfully obtained heteroepitaxial growth of a few nanometer-thick sp^2 hybridized BN film of well-stacked and flat layers on Ni (111) surface of polycrystalline substrate [4]. Here, we will present our work on Rapid Thermal CVD from Annealsys (www.annealsys.com). We will show how we successfully adapt our growth process to this new CVD reactor on centimeter monocrystalline nickel (111) substrates. We will detail the crucial step of nickel surface preparation before the synthesis. We will present the results of the structural and quality characterization of the BN films from the macroscale to the nanoscale (OM, SEM, TEM, AFM, Raman spectroscopy) on the growth substrate and after transfer onto dedicated substrate

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Mass Production of Two-Dimensional Materials by Intermediate-Assisted Grinding Exfoliation

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Two-dimensional (2D) materials with intriguing physical and chemical properties are expected to have a variety of applications. However, the scalable and high-efficient production of 2D materials with a large aspect ratio and high quality is still a challenge. Our group has developed a series of exfoliation methods that use micro-particles or polymers as force intermediates to assist the exfoliation of layered materials, which are referred to as (polymer) intermediate-assisted grinding exfoliation, with the abbreviation of (p)-iMAGE. With micro-particles assistance, we prepared 2D h-BN with a high yield of 67%, a high production rate of 0.3 g $h^{-1,}$ and a low energy consumption of 3.01×10^6 J g^{-1}. In this method, micro-particles like SiC increases the friction coefficient of the mixture and convert the applied compressive forces into a multitude of small shear forces exerted on layered materials, promoting exfoliation efficiency. With polymers assistance, we produced 2D h-BN with an average lateral size of 2.18 μm and thickness of 3.91 nm. In this method, polymers like CMC have bi-functions as intercalant and adhesive agents, which not only buffer against high energy input from pestle but also realize one-step fabrication of composites, more convenient for the following applications like thermal management. These methods are also universal in exfoliating other 2D materials. Our works indicate the huge potential to produce large amounts of various 2D materials, which paves the way for their commercial applications.

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Epitaxial growth of single-crystal hexagonal boron nitride multilayers

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Large-area single-crystal monolayers of two dimensional (2D) materials such as graphene, hexagonal boron nitride (hBN) and transition metal dichalcogenides have been successfully grown. Among them, hBN has been demonstrated to be the "ideal" dielectric substrate for 2D materials-based field effect transistors (FETs) – offering the potential for extending Moore's law. Although hBN thicker than a monolayer is more desirable as substrate for 2D semiconductors, the growth of highly uniform and single-crystal few- or multi-layer hBN has not yet been demonstrated. Here we report the epitaxial growth of wafer-scale single-crystal tri-layer hBN by a chemical vapour deposition method. Uniformly aligned tri-layer hBN islands are found to grow on a 2 cm × 5 cm single-crystal Ni (111) at early stage of growth and finally to coalesce into a single-crystal film. Cross-sectional transmission electron microscopy (TEM) results show that a $Ni_{23}B_6$ interlayer is formed (during cooling) between the single-crystal tri-layer hBN film and Ni (111) substrate by boron dissolved in Ni (111) and that there is epitaxial relationship between tri-layer hBN and $Ni_{23}B_6$ and between $Ni_{23}B_6$ and Ni (111). We further find that the tri-layer hBN film acts as a protective layer that remains intact during catalytic evolution of hydrogen – suggesting continuous and uniform single-crystal tri-layer hBN in large area. This tri-layer hBN transferred onto the SiO_2 (300 nm)/Si wafer acts as a dielectric layer to reduce electron doping from the SiO_2 substrate in MoS_2 FETs. Our results demonstrate that it is possible to achieve high quality multi-layered hBN over large areas by CVD – opening up new pathways for making it a ubiquitous substrate for 2D semiconductors and other purposes.

Design of satellite-based hBN single-photon sources for quantum communication

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In recent years, quantum technologies have advanced significantly, enabling their use in space-based applications such as long-distance quantum communication. In this paper, we present the design of a compact true single photon source based on a fluorescent color center in hexagonal boron nitride. This innovative light source offers improved secure data rates in satellite-based quantum key distribution scenarios compared to conventional laser-based light sources. The emitter is off-resonantly excited by a diode laser and coupled to an integrated optical circuit that routes photons to different experiments performed directly on-chip. These experiments either characterize the single photon source using the second-order correlation function or test a fundamental postulate of quantum mechanics, Born's rule, which relates the probability density and the wave function. Our payload is currently being integrated into a 3U CubeSat and scheduled for launch in 2024 into low Earth orbit. Thus, we can evaluate the feasibility of true single-photon sources in space and provide a promising route toward a high-speed quantum.

Epitaxial growth and anti-corrosion behavior of two-dimensional hBN on copper

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Recently, the rising of two-dimensional (2D) materials brought the significant development of several cutting-edge technologies. Among the 2D family, hexagonal boron nitride (hBN) is found to be an essential one, due to its exclusive wide band gap (about 6 eV) and free of dangling bond. The industrial-level applications require massive preparation of large-size, high-quality film. In this talk, I will introduce our recent progress on the fabrication of A4-paper sized single-crystal Cu foil library[1], the epitaxial growth of 100-square-centimetre single-crystal hBN monolayer on as-prepared single-crystal Cu foil[2], and the abnormal anti-corrosion behavior of hBN grown on Cu foil[3].

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Biograph:

Kaihui Liu is currently a professor of Physics in Peking University, China. He received his Ph.D. from the Institute of Physics, Chinese Academy of Sciences in 2009 and afterward worked as a postdoctoral research fellow at UC Berkeley until 2014. His current research interests are the manufacture, characterization and application of two-dimensional (2D) single-crystal materials. His main academic achievements include (i) designing the interfacial element feeding for the ultrafast and low-defect growth of 2D materials, (ii) developing a universal method for the synthesis of meter-sized 2D single-crystals, and (iii) fabricating advanced 2D materials-optical fibre devices. Currently, Prof. Liu serves as the Associate Editor of Science Bulletin and project leader of National Key Program of China. Besides, he has received several honors, including the Xplorer prize (2021), National Science Fund for Distinguished Young Scholars (2020), Beijing Natural Science Foundation for Distinguished Young Scholars (2019), Beijing Outstanding Young People (2017), and Ten "Xin Rui" Scholar of China (2016).

Growth of Distributed Bragg Reflectors entirely made of boron nitride

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Distributed Bragg Reflectors (DBRs) are one of the major building blocks of many optoelectronic devices. Although epitaxial techniques like Metal-Organic Chemical Vapor Deposition (MOCVD) have been used to fabricate DBR structures for more than three decades, utilizing boron based compounds is a relatively new idea [1,2]. Commonly, the required difference in the refractive index values between the two component layers of the DBR is obtained by using two different materials. However, the refractive index contrast may also be achieved via introducing porosity to at least one component layer [3,4].

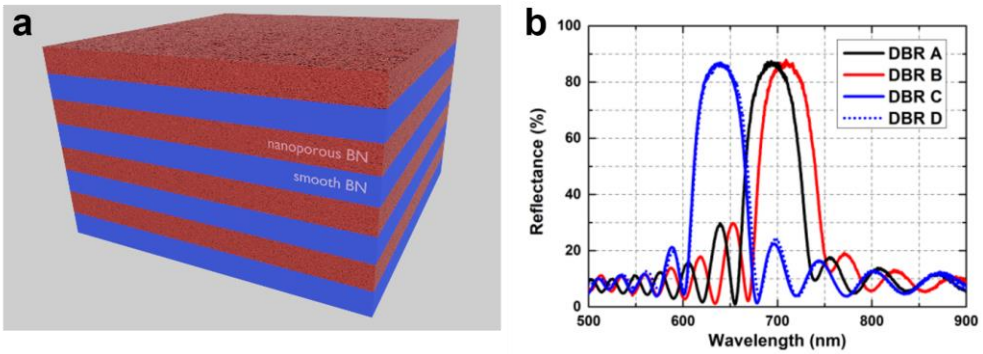

Fig. 1: (a) Schematic illustration of a DBR based solely on BN. (b) post-process reflectance spectra of the fabricated DBR structures,

In this communication, we show the fabrication and optical performance of DBRs for which both component layers are made of BN, but exhibit varying levels of porosity and therefore have different values of the refractive index (Fig.1a). We are able to achieve this by manipulating only the substrate temperature during the growth of the sample [5]. In contrast to the case of porous DBRs based on other material systems [3,4], the fabrication of the whole structure is possible in a single MOCVD process, without the need for any post-process etching. For DBRs consisting of 15.5 layer pairs, we were able to achieve a peak reflectance reaching 90% (Fig 1b) [5]. Moreover, we were able to demonstrate an air-filled optical microcavity based on the fabricated DBRs. This opens up possibilities for all-BN single photon sources, for which hBN-based single-photon emitters would be embedded within the cavity of BN-based DBRs, being fabricated in a single MOCVD process.

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ZnO nanorods pre-orientated by hexagonal boron nitride on copper paper for multiple applications

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Growth of high quality semiconductor crytal on polycrystalline or amorphous substrates is a long-term challenge. We found that hexagonal in-plane lattice arrangements and weakly bonded layers of hexagonal boron nitride (h-BN) could be useful for relaxing the lattice and thermal mismatches between substrates and heteroepitaxial crystal. Thus, 2D h-BN monolayer can be used as a pre-orientation layer to induce van-de-Waals growth of semiconductor on arbitary substrates.

In our work, direct epitaxy of ZnO nanorods array was carried out on polycrystalline Cu paper by employing h-BN as a pre-orientating interlayer. Single-crystal and vertically aligned ZnO nanorods array could be obtained in large length (~ 75 μm) through chemical vapor deposition (CVD) or hydrothermal methods. In addition, the h-BN monolayer meanwhile is an excellent dielectric layer for device fabrication. The extreme effect when the dielectric thickness reaches the limit of atomic monolayer can greatly improve the performance of devices.

For optoelectronic applications, we fabricated (1) a Cu/h-BN/ZnO/Cu NWs flexible solar-blind photodetecting paper, which reveals the ultra-high responsivity up to 700 A/W @ 265 nm and high photoconductive gain of ~ 2×10^3 [1]. (2) The h-BN/ZnO nanorods/h-BN sandwiched strucutre was designed for fabricating transparent and flexible piezoelectric nanogenerator (PENG), which showed the open-voltage of 5 V, short-current of ~ 18 μA, maximum output power density up to 169 mW/cm^2 [2].

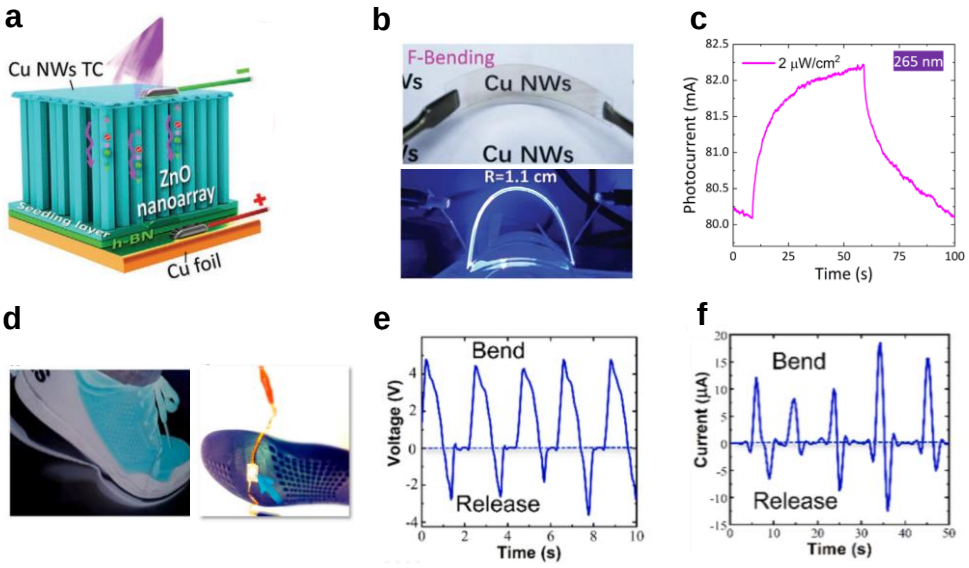

Figure 1. (a)-(c) ZnO nanorods pr-orientated on Cu foil by h-BN for solar blind photodetecting paper. (d)-(f) PENG film device fabricated with h-BN/ZnO nanorods/h-BN sandwiched structure for harvesting body movement energy.

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Machine Learning Assisted Calculation Of Phonon Properties In Layered hBN

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The calculation of finite temperature properties of solids is a topic under heavy discussion in recent years. There are mainly three predominant approaches to the problem: purely harmonic theory [1], effective harmonic models (e.g. the sSCHA [2] or sTDEP [3,4]) and *ab initio* molecular dynamics (aiMD) simulations [5].
The latter two overcome the clear faults of the first by correctly predicting quantities like thermal expansion and the shift of the potential energy surface minima.
They do, however, suffer from the same problem of needing large amounts of DFT calculations in order to obtain converged results. This is especially the case for aiMD, where some systems reach prohibitive calculation times to obtain relevant properties.
In this work, we show how taking advantage of the ML assisted canonical sampling (MLACS) and interatomic potentials (MLIP) [6] as a general accelerating method, allows for the usage of long propagation time MD and TDEP as an easy method for predicting accurate temperature dependent quantities. We examplify this with the cases of monolayer and bilayer hexagonal boron-nitride (hBN) by calculating the temperature evolution of their phonon band-structures, Raman shifts and thermal conductivities.

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Single photon emitters in hBN via ultra-low energy helium ion implantation

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A discovery of quantum emitters in hexagonal boron nitride (hBN) has recently incited immense interest for quantum technologies. It offers a platform for fundamental science but is of interest for applications in quantum photonics owing to its robust single photon emission at room temperature. Recent studies have suggested that these SPEs are associated with intrinsic defects, which led to efforts to engineer the SPE in hBN by various methods such as plasma treatment, annealing, laser, e-beam and ion irradiation methods. Despite these efforts, the origin of single photon emission and the correlation of emission with particular defects still need to be scrutinized. Here we use ultra-low energy ion implantation to introduce defects in hBN. We show that helium ions with energies as low as 50 eV are extremely efficient in introducing single photon emitters in hBN. We also show that low temperature annealing increases the density of the emitters. We consider the possible defects that helium ions at the implantation energy can generate in hBN and use statistical data on single photon emitters to discuss the possible origin of the emission. Finally, we discuss the viability of creating emitters in pre-selected locations.

Fingerprinting color centers in hexagonal boron nitride for quantum technology application

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Abstract

Optical quantum technologies have the potential to revolutionize today's information processing and sensors. Crucial to many quantum applications are efficient sources of pure single photons with the optical emission wavelength being application-specific. Here, we employ density functional theory (DFT) to reveal the photophysical properties of the single photon emitters in two-dimensional hexagonal boron nitride. Among 267 different defects characterized by the HSE06 functional, we select promising defects suitable for direct coupling to other quantum systems and applications in quantum technologies. Then, the photoluminescence spectrum, lifetime, quantum efficiency, and dipole transition of the selected defects are also unraveled. This work thereby provides the entire fingerprint of quantum emitters, which is a promising way to identify hBN emitters instead of relying on only a single property such as the photoluminescence spectrum. In addition, we also provide a recipe on the generation of a universal solid-state quantum emitter system for optical quantum technologies.

First-principles study of luminescence in hexagonal boron nitride single layer: exciton-phonon coupling and the role of substrate

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Hexagonal boron nitride (hBN) is a wide band gap material with both strong excitonic light emission in the ultraviolet and strong exciton-phonon coupling. Recent luminescence experiments performed on the synthesized monolayer form (m-hBN) present emission spectra that differ from one another, with some suggesting a coexistence between phonon-assisted and direct emission channels [1-3]. Motivated by these results, we investigated the optical response of (m-hBN) using a new *ab initio* approach that takes into account the effects of atomic vibrations on the luminescence spectra. We construct the dynamical exciton-phonon self-energy [4], then use it to perturbatively correct the optical response functions and test this approach on bulk hBN as a benchmark. Within our approach we are able to estimate the renormalisation of the direct peak induced by phonon-assisted transitions, and this allows us to accurately describe spectra where both processes are present. As the experiments were performed with different substrates, we also investigate the effect of a graphite substrate on the photoluminescence spectrum.

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Toward non-gas-permeable hBN film growth on smooth Fe surface

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Hexagonal boron nitride (hBN) has been highlighted due to its high optical transparency in the visible range and high-temperature oxidation resistance up to 1500 ℃ in air [1]. Typically, large-area m-hBN films have been synthesized on Fe-based substrates due to reasonable B and N solubilities in Fe bulk [2-4]. In this precipitation stage, rough Fe surfaces composed of atomic grooves, steps, and kinks on a Fe grain in polycrystalline Fe foil produce a non-uniform and discontinuous polycrystalline hBN film [2, 3]. Thus, an atomically flat surface is desirable to achieve a high-quality m-hBN film.

In this study, we report an effective growth method to improve the quality of m-hBN in terms of its uniformity and continuity by using a smooth Fe surface. The smooth Fe surface allows the uniform precipitation of B and N to grow highly uniform and continuous m-hBN films. Further, the m-hBN/Gr heterostructure film exhibits impressive WVTR performance with high transparency compared to previously reported values.

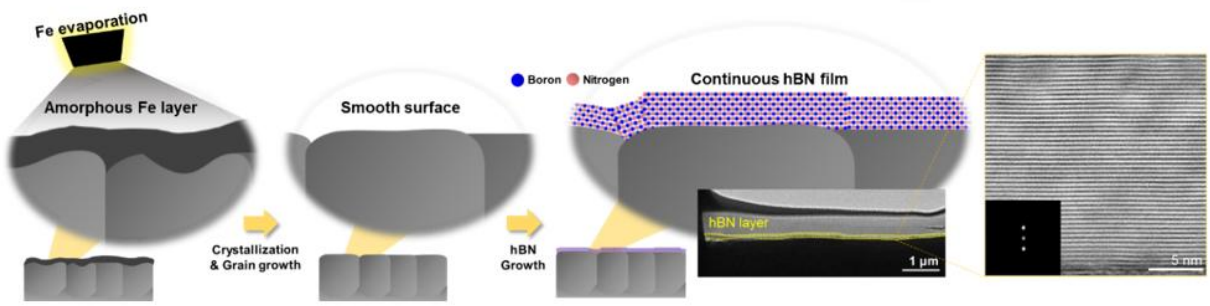

Fig.1 Schematic illustration of this concept and the cross-sectional TEM images.

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Extending the coherence time of spin defects in hBN enables advanced qubit control and quantum sensing[1]

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In this work, we address the problem of the short spin coherence time of the VB^- defects in hBN, a severe limitation for their application in quantum technology.
Specifically, we apply dynamical decoupling techniques to extend the spin coherence time of VB^--ensembles in hBN by nearly two orders of magnitude, approaching the fundamental T_1 relaxation limit. The improved coherence times enable us to demonstrate the detection of AC magnetic fields in several complementary experiments, including sensing RF signals with sub-Hz frequency resolution.
The presented work broadens the applicability of VB^- defects in hBN for nanoscale quantum technologies and paves the way to establish magnetic resonance sensing in ultra-thin structures.

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Coherent dynamics of multi-spin V_B^- center in hexagonal boron nitride

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Hexagonal boron nitride (hBN) has recently been demonstrated to contain optically polarized and detected electron spins that can be utilized for implementing qubits and quantum sensors in nanolayered-devices. Understanding the coherent dynamics of microwave driven spins in hBN is of crucial importance for advancing these emerging new technologies. Here, we demonstrate and study the Rabi oscillation and related dynamical phenomena of the negatively charged boron vacancy (V_B^-) spins in hBN. We report on different dynamics of the V_B^- spins at weak and strong magnetic fields. In the former case the defect behaves like a single electron spin system, while in the latter case it behaves like a multi-spin system exhibiting the multiple-frequency dynamical oscillation like clear beat in Ramsey fringes. We also carry out theoretical simulations for the spin dynamics of V_B^- and reveal that the nuclear spins can be driven via the strong electric-nuclear coupling existing in V_B^- center, which can be modulated by the magnetic field and microwave field.

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Emitter-Optomechanical Interaction in Ultra-High-Q hBN Nanocavities

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Hexagonal boron nitride (hBN) is a wide bandgap 2D semiconductor that has recently been found to host a variety of point defects of interest for single photon generation, spin-photon interface, and quantum sensing. The negatively charged boron vacancy V_B^- attracts particular interest due to its paramagnetic nature and spin-dependent optical properties. V_B^- is a unique emitter of which the emission is dominated by phonon-induced processes. The emission dynamics and zero-phonon energy of V_B^- are not well known yet. We unveil the zero-phonon energy of V_B^- by the cavity enhanced emission [1]. Furthermore, the dominance of phonon-induced processes indicates that V_B^- is intrinsically sensitive to and interacts with the local phonon mode induced by lattice phonons and cavity optomechanical vibrations. We probe the phonon processes in the V_B^- emission and show how the V_B^- related phonons bridge the coupling between V_B^- emission, cavity photons and nanomechanical cavity vibrations, establishing the strong interplay between multiple degrees of freedom in the emitter-optomechanical system [2]. Such multi-modal couplings provide provides novel methods to interface spin defects, photons, and phonons in condensed matter systems.

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Quantum emitters are needed for a myriad of applications ranging from quantum sensing to quantum computing. Hexagonal boron nitride (hBN) quantum emitters are one of the most promising solid-state platforms to date due to their high brightness and stability and the possibility of a spin − photon interface. However, the understanding of the physical origins of the single-photon emitters (SPEs) is still limited. Here we report dense SPEs in hBN across the entire visible spectrum and present evidence that most of these SPEs can be well explained by donor − acceptor pairs (DAPs). On the basis of the DAP transition generation mechanism, we calculated their wavelength fingerprint, matching well with the experimentally observed photoluminescence spectrum. Our work serves as a step forward for the physical understanding of SPEs in hBN and their applications in quantum technologies.

Improving optical response of layered semiconductors via hBN encapsulation

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Hexagonal Boron Nitride (hBN) played a pivotal role in the development of van der Waals heterostructures and devices, as an excellent dielectric to fabricate the gates, as well as an atomically smooth substrate and encapsulating milieu, protecting against degradation and contamination with ambient agents. Many of 2D materials are semiconducting, making them candidates for opto-electronic applications. The optical response of such materials, as MoS_2 [1], $MoSe_2$ [2, 3], WSe_2 [2] or CrSBr [4], is dominated by excitons of a particularly high binding energy beyond 100 meV and strong optical absorption of around 10%. Due to the apparent strong light-matter interaction, the optical nonlinearities are enhanced, which can be probed via nonlinear micro-spectroscopy.

In this contribution, by measuring coherent response of excitons, namely their four-wave mixing polarization, I will show the universal improvement of optical properties of 2D materials via hBN encapsulation. In top-down (exfoliated) MoS_2, $MoSe_2$, WSe_2, and CrSBr heterostructures, it permits as to suppress the disorder to the point where the spectral line shape is dominated by a homogeneous broadening. In a bottom-up approach the successful epitaxial growth, yielding $MoSe_2$ monolayers of a good optical quality, is enabled by employing atomically flat hBN substrates [5, 6]. hBN hosts a plethora of defects acting as quantum emitters operating up to ambient conditions. It is therefore a promising material for quantum optics applications beyond cryogenic temperatures, which in a near future could be efficiently probed with four-wave mixing microscopy.

The hBN employed in these studies was provided by the **partners from NIMS in Tsukuba**, whereas the heterostrucutres were fabricated at **TU Munich**, **LNCMI & Institut Néel CNRS Grenoble**, **University of Manchester** and **University of Warsaw**.

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