Research Statement and Plans: Abdelghani Laraoui, CHTM-UNM

My research interests lie at the intersection of experimental condensed matter physics with quantum optics and materials science. My approach integrates concepts from quantum optics and methods from condensed-matter nanotechnology to tackle outstanding challenges in metrology, computing, and physical science. I envision starting a multidisciplinary laboratory composed of and collaborating with condensed-matter and bio physicists, and other faculty members at Texas Tech University. The several lines of my research which I will pursue are: I) study novel solid-state materials for quantum information processing, optoelectronics, and spintronics applications, II) Nanoscale mapping of phenomena in condensed matter physics using solid-state-quantum sensors. Basic and advanced analytical and spectroscopic tools will be used, such magneto-optical spectroscopy (Kerr, Faraday), scanning probe microscopy (AFM, MFM, etc.), electron microscopy (TEM, STM), magnetometers (VSM, SQUID), magneto-transport, and optical spectroscopy (Raman, ultrafast pump-probe, fluorescence). Basic materials coating, optical lithography, and thin film deposition (sputtering, MBE, etc.) will be used, and external collaboration will be established to get advanced materials.

Recent/current research: My current research plans are centered around using color centers in diamond based on nitrogen-vacancy (NV) centers. NV centers are presently the focus of a broad cross disciplinary research effort combining the fields of condensed matter, quantum physics, and precision metrology. Several unique properties, including the long spin coherence times exceeding one millisecond and superb photostability at room temperature [1], make them central to various proposals for quantum information processing (QIP) and nanoscale metrology [2]. Applications facilitated by the ability to initialize, manipulate, and readout spins optically with high fidelity from cryogenic environments to room temperature. At City College of New York, I used NV centers as probes for high-resolution magnetic and temperature sensing. I demonstrated that it is possible to use an NV center confined within a diamond nanocrystal to probe the surrounding paramagnetic and surface spins for enhanced biomolecular imaging [3]. I introduced a new spin-correlation protocol for implementing nuclear spins (^{14}N, ^{13}C) as quantum registers [4], and developed a novel approach of thermal conductivity and temperature imaging based on scanning spin probe for sensitive nanoscale *thermometry* [5]. Currently at University of New Mexico I am developing new probes based on NVs, silicon vacancy centers (SiV) in diamond and defects in wide bandgap semiconductors for quantum sensing. I am also developing new microscale/nanoscale magnetic resonance protocols [6] based on nanophotonics, studying malarial hemozoin biocrystals [7], and exploring magnonic crystals and multiferroics for coupling distant spin-qubits for scalable quantum networks [8].

Research integration of graduate and undergraduate students:

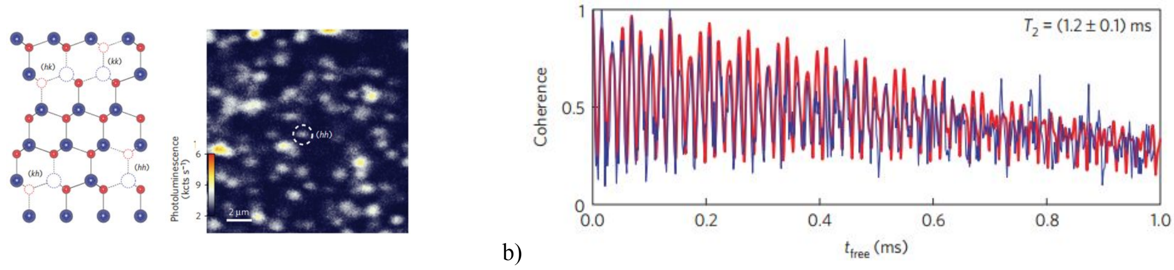
My research program expects to accommodate students with diverse research interests, allowing a multi-layered structure to accomplish the research objectives. I will undertake flexible and teamwork-based strategies, so that undergraduate and graduate students of Physics & Astronomy Department and other TTU departments can balance their core coursework and achieve publishable results. This way, students at different academic levels can participate and collaborate, appreciating the importance of their research goals and specific perspectives. My research approaches will offer multifaceted avenues to augment undergraduate and graduate students' professional interests and practical skills, providing opportunities for experimental/theoretical studies, and engineering projects. I will design well-rounded summer research plans and encourage undergraduate students' participation in scientific conferences. I will seek extramural funding from federal organizations, private foundations and industrial partners to energize my research program. I will strive to build up collaborative projects with faculty members at the Physics Department and with larger research community in the university to foster new interdisciplinary research avenues. Together with a strong commitment to undergraduate and graduate teaching, my goals rely on strengthening a multi-layered, interdisciplinary research program through a comprehensive mentoring initiative for diverse student ensembles at TTU.

I- NOVEL SOLID-STATE MATERIALS FOR QUANTUM INFORMATION PROCESSING, OPTOELECTRONICS, AND SPINTRONICS APPLICATIONS

I will characterize selective quantum grade quality novel host materials wide-band-gap (WBG) materials (e.g. ZnO, AlN) and two dimensional (2D) materials (e.g. h-BN, WSe_2, MoS_2) with desired color centers

exhibiting unique quantum properties. This will elucidate the physical mechanisms responsible for the observed novel quantum properties (e.g. spin coherence) and governing composition- processing defect-property relationships. My group will also develop new characterization techniques tailored to varieties of excitations (electrical, magnetic, thermal, mechanical) and sensing techniques. Long term, I envision application for innovative device designs and sensors based on their novel properties. I anticipate forming collaborations with material scientists, optics researchers and solid-state physicists at TTU and worldwide. We will seek funding from NSF (quantum materials, spintronics), DARPA, ARL and AFOSR.

a)



b)

Figure 1. Quantum defects in WBG semiconductors. a) left. Divacancies in 4H–SiC consist of neighbouring Si and C vacancies. Because either the h or k lattice site can be vacant, there are four inequivalent forms of divacancy in 4H–SiC. a) right. confocal photoluminescence image from a 4H–SiC membrane. b) spin coherence of a (kk)-divacancy ensemble in SiC exceeding 1 ms at ambient conditions [10].

This project is centered around exploring ***novel solid-state*** materials and spin-qubits of ***color centers*** and ***defects*** in wide bandgap semiconductors for quantum information science, nanoscale metrology, and biosensing. Although significant progress has been achieved in understanding and utilizing the quantum properties of optically addressable NV centers in diamond for quantum sensing and quantum computing, further advances are severely limited by difficulties in achieving exact placement of NV centers, light collection due to the high refractive index of diamond, large scale integration, and low qubit yield. Superior solid-state host materials such as 3D WBG semiconductors and recently discovered atomically thin 2D van der Waals materials with varieties of optically addressable color centers are very attractive alternatives to advance this science. **The scientific aims and timeframe of this project are the following:**

Aim 1 (section Ia, years 1-3). My group will study different defects in WBG semiconductors such ZnO and SiC. Later other materials can be explored such AIN and GaN. This part of the project aims to: ***i)*** measure the spin coherence life times of the defects, ***ii)*** explore single photon emission for integration to optoelectronic devices, ***iii)*** develop new characterization techniques tailored to varieties of excitations (optical, electrical, magnetic, thermal, strain, etc.) and sensing techniques. To perform such study my group will build a custom cryogenic confocal fluorescence microscope with a continuum laser excitation integrated to a spectrometer and an optical detected magnetic resonance (ODMR) setup.

Aim 2 (section Ib, years 3-5). My research goals of this part include: ***i)*** characterize selective quantum grade quality novel host 2D materials (e.g. h-BN, WSe_2, MoS_2) with desired color centers exhibiting unique quantum properties, ***ii)*** Elucidate the physical mechanisms responsible for the observed novel quantum properties (e.g. spin coherence) and governing composition- processing defect- property relationships, ***iii)*** Explore innovative device designs based on the qubits novel properties.

I.a Defects spectroscopy in WBG

WBG materials offer varieties of color and paramagnetic centers with narrow tunable spectral emissions and long coherence spin times as well as allow integration with nano-photonics and spintronics possible. Identifying defects with similar NV properties would add flexibility in device design and possibly lead to superior performance or greater functionality. A systematic search for defect-based qubits has been initiated [9], starting from a list of physical criteria of such centers and their hosts should satisfy:

- a wide bandgap (eg: diamond (= 5.5 eV), SiC (= 2.2- 3.3 eV), ZnO (= 3.3 eV)), so that it can accommodate a deep center: the optical transitions do not introduce interference from the electronic states of the host.
- a small spin-orbit coupling, in order to avoid unwanted spin flips in the defect bound states.
- availability as high-quality, bulk, or thin-film single crystals, in order to avoid imperfections or paramagnetic impurities that could affect the deep center's spin state.
- Constituent elements with naturally occurring isotopes of zero nuclear spin, so that spin bath effects may be eliminated from the host via isotopic engineering.

Defects of a potential interest includes: deep centers in high-purity monocrystalline 4H–Silicon carbide (SiC) with exceptionally long coherence times exceeding one millisecond at room temperature [10] (**Figs. 1. a), b)**); color center in ZnO [11], point defects and defect complexes in AlN for potential applicability as single-spin centers and solid-state qubits [12]. By exploiting spin-dependent optical transitions, ODMR, optical spectroscopy, and microscopy (SEM, TEM) will be used for achieving single-spin addressability in the solid state and exploring new defects. The quantum-optical properties of these quantum systems will be studied at temperatures varying from 4 to 300 K. Furthermore, since the number of emitters in these nanoscale materials is small (down to just a single emitter), the measurement apparatus must combine several state-of-the-art specifications:

1. single-emitter detection sensitivity; individual photons must be detected with high efficiency.
2. single-emitter addressability: the laser optical frequency must be stabilized to within the emitters' narrowest spectroscopic features, <1 MHz.
3. high spectroscopic sensitivity; the emission optical frequencies must be determined to better than a part per million, ~100 MHz.
4. nanometer mechanical control; in order for devices to be aligned to free-space optical components, mechanical control and stabilization at the few nm level is required.

Fig. 2a depicts a typical configuration of a cryogenic microscope used to study silicon vacancy (SiV) centers in diamond at UNM. The proposed microscope uses widely tunable lasers suitable for defects spectroscopy in WBG, a more optics-friendly cryostat design, and higher-resolution spectroscopy.

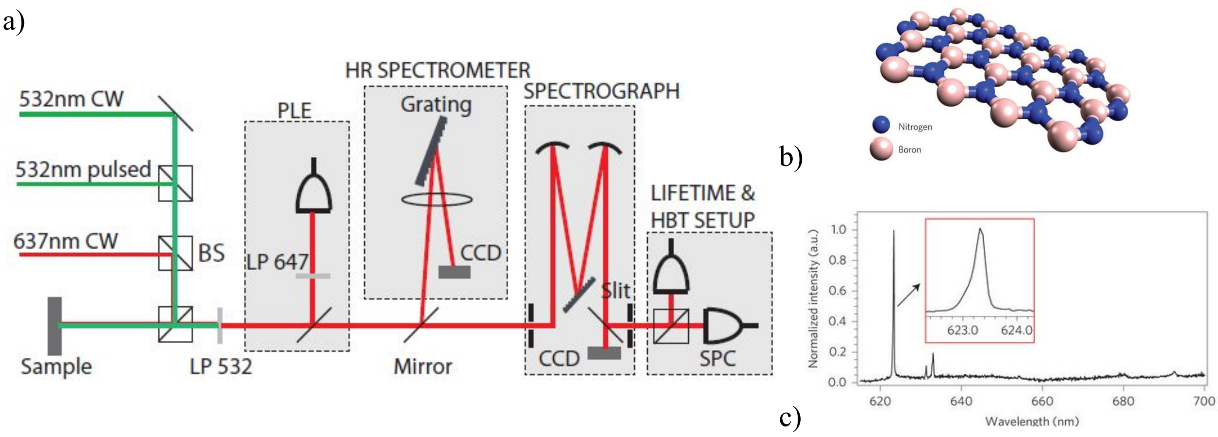


Figure 2: Experimental configuration. (a) cryogenic microscope used to study SiV centers. The emitters are addressed using 532 nm and 637 nm lasers. Photoluminescence excitation (PLE) spectroscopy is performed by monitoring emission of off-resonant single photons, sending resonant emission to a high-resolution home-built spectrometer (HR Spectrometer), sending emission through a lower-resolution commercial spectrometer (spectrograph), and/or using the spectrograph to filter emission before performing quantum optical analysis such as emitter lifetime and autocorrelation analysis. (b) Schematic of the hBN lattice structure. (c) PL spectrum taken at 77 K of a defect center in multilayer hBN. Inset: the zero phonon line [20].

I.b Individual color centers in 2D materials

Atomically-thin materials have attracted tremendous interest for their bulk "2D" properties including mechanical strength, electronic transport, electrically-tunable band structure, and extraordinary optical confinement. They have enabled new technological breakthroughs across a variety of disciplines in materials science, nanophotonics and physics, including: realization of phonon polaritons [13], lasing [14], and spin valley transport [15]. It is natural to wonder whether the perfect color center is one which resides in such a 2D material yet has the ideal spin and optical properties of defects in bulk semiconductors such as diamond NV and SiV centers, etc. The 2D host might facilitate enhanced defect properties such as:

1. A color center in a 2D material would exhibit higher photon collection efficiency; the 2D host does not suffer from reflections at index mismatches as its 3D counterpart does.
2. The ability to inject carriers by applying large electric fields to, for example, excite electroluminescence or stabilize a defect against (de)ionization is easier in a 2D material, compared to bulk materials which require p/n doping [16].

3. Optical cavities with extremely-small mode volumes are relatively straightforward to fabricate with a 2D material. In such a cavity, a color center with a broad emission band could still exhibit cavity-enhanced emission via the Purcell effect [17].
4. 2D materials offer unique mechanical properties including exceptional stiffness, which allows for mechanical oscillators mechanical cavities with high quality factor and high resonance frequency. A color center embedded in such a resonator could enable a new regime in optomechanics such as room-temperature ground-state cooling [18].
5. An array of color centers in a 2D material offers an ideal geometry for many-body quantum simulations, a potential room-temperature solid-state alternative to atoms in optical lattices.

While defects in 2D materials have been known to exist for several years, it wasn't until 2015, that scientists managed to isolate individual color centers in these systems. Several groups reported observation of defects in WSe2 that exhibit single photon emission at low temperature [19]. Later, a single-photon emission from localized defects in hexagonal boron nitride (h-BN) monolayers have been observed at room temperature [20] (**Figs. 2b, 2c)**), enabling the transformational application of 2D materials in quantum information applications with the potential to achieve scalable nanophotonic circuits on a single chip. Thus, this project aims to identify a suitable analogous system to the diamond NV centers for a myriad of applications in sensing and computing. This include: synthesis of isotope free quantum grade novel host materials, identification and incorporation of unique color centers, characterization of their quantum properties (spin coherence time, spectral stability) and the understanding of their interactions with the external excitations (optical, electrical, magnetic).

II. NANOSCALE MAPPING OF PHENOMENA IN CONDENSED MATTER SYSTEMS USING NV MAGNETOMETER

I will approach the study of technologically-relevant materials from a new angle, with ambitious goals such as elucidating the mechanisms of high-Tc superconductivity in the hunt for room-temperature superconductivity, exploring the magnetic properties of novel magnetic nanomaterials like skyrmion states in thin ferromagnetic films, and mapping the spin current generated in 2D topological insulators for data storage and spintronics applications. I will build magneto-optical NV-based microscopes that will be indispensable tools for nanomaterials laboratories by offering massively parallel operation over a wide temperature and magnetic field ranges. I intend to collaborate with condensed matter physicists and spintronic physicists, and seek funding from NSF, ONR, ARL, and other funding agencies.

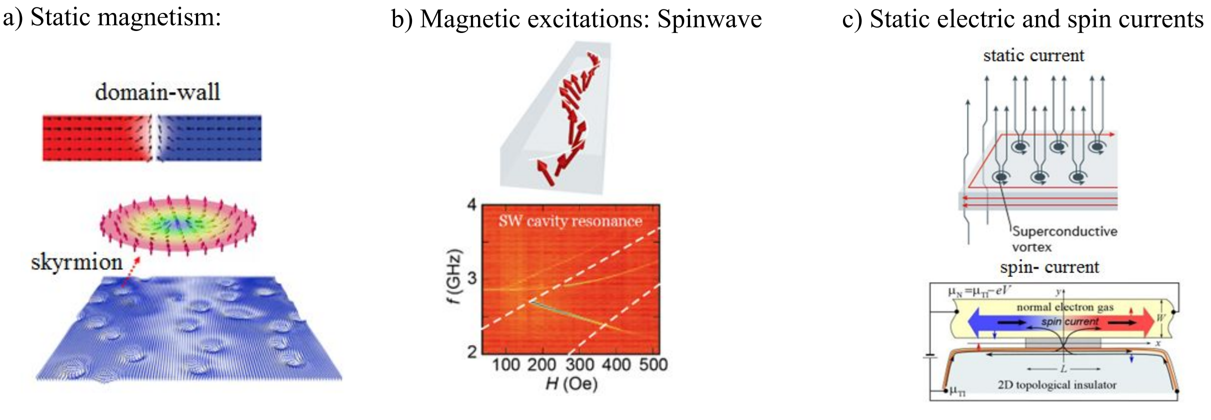


Figure 3. Nanoscale mapping of phenomena in condensed matter physics (a) static magnetism such domain-wall in thin ferromagnetic thin films (upper) or 2D skyrmions states (lower) micromagnetic in a ferromagnetic thin film [21]. b) Magnetic dynamic excitations such spin-wave measured using NV center in YIG thin film (lower) [22]. c) Static current generated by superconductive vortices [24] (upper) and spin current generated in topological insulators (lower) [23].

Understanding the behaviour of spins and charges in materials is at the heart of condensed matter physics. In the past few decades, a wide range of new materials displaying exciting physical phenomena has been discovered and explored. Examples include van der Waals materials [25], topological insulators [26] and complex oxide interfaces [27]. There is intense ongoing activity focused on developing and understanding these materials and on creating new ones. The success of these efforts relies on advances in theory and materials synthesis and on the development of sensitive measurement techniques. Because spins and moving charges generate stray magnetic fields, a local and non-perturbative magnetic field sensor that can operate over a broad temperature range could be used to characterize the growing number of correlated and topological electron systems. Spin and charge sensing at the atomic scale would have wide-ranging applications, including: ***(i)*** determining the static magnetic configuration in thin ferromagnetic/antiferromagnetic thin films and nanomagnets with novel functionalities such domain-wall and skyrmion states (**Fig. 3a**), ***(ii)*** measuring the dynamics of magnetic excitations in magnetic insulators and nanomaterials (**Fig. 3b**), and ***(iii)*** probing the static charge and spin currents generated in superconductors and 2D topological insulators (**Fig. 3c**). The spin of an elementary particle such as an electron or a nucleus can be used as an atomic-scale magnetic field sensor. Thus, spin-based magnetometry techniques, such nuclear magnetic resonance (NMR) and neutron scattering, give access to the magnetic structure of a material on the atomic scale. However, these techniques do not provide real-space imaging or sensitivity to samples with nanoscale volumes. By contrast techniques such as magneto-optical Kerr microscopy (MOKE) [28], magnetic force microscopy (MFM), magnetic resonance force microscopy (MRFM) [29], and scanning superconducting quantum interference devices (SQUIDs) allow real-space imaging of the magnetic fields emanating from nanoscale devices, but they have a finite size and act as perturbative probes and/or over a narrow temperature range. Magnetometry based on the electron spin associated with the NV center in diamond (**Fig. 4**) combines powerful aspects of both modalities. The NV spin is an atomic-sized sensor that benefits from a large toolbox of spin manipulation techniques and can be controllably positioned within a few nanometres of the system under study. However, only in the past few years has NV magnetometry begun to be used to explore condensed matter systems. **The scientific aims** of this project are the following:

Aim 1 (years 1-2, section IIa). My group will build magneto-optical NV based microscopes in near field, i.e., a confocal microscope integrated to scanning probe microscope, and in far-field. To test the sensitivity of the microscopes, my group will study the static and magnetic dynamic properties of individual **transition metallic magnetic nanoparticles (MNPs)** with 2-10 nm diameter around the superparamagnetic-ferromagnetic transition (~4 nm for Co nanoparticles) [30]. These particle are promising candidates for high-density magnetic recording [31], but their small size has so far precluded studying their magnetic dynamics at the individual particle level. Using diamond magnetic microscopy, we seek to establish a fundamental understanding of the effect of size, surface structure, and inter-particle dipolar interactions on their magnetic properties.

Aim 2 (years 2-5, section IIb, IIc). Using the NV magnetometer, my group will study **low dimensional spin systems** based on topological states in thin/nanostructured magnetic films and **surface spin currents generated in 2D topological insulators** [23], **Fig. 3c)**. These spin systems have attracted a huge interest the last few years due to their novel properties and integration in spintronics and magnetic devices. Other interesting systems to study include, **magnetic skyrmions (Fig. 3a)), spin-textures of multiferroic** thin films, and **dynamic magnetic excitation** (spinwave), **Fig. 3b)** [21].

II.a NV magnetic microscopy

Recently, a new technique has emerged for measuring magnetic fields at the nanometer scale based on optical detection of the electron spin resonances of NV centers in diamond, **Figs. 4a), 4b)**. Negatively charged NV centers, comprised of a substitutional nitrogen adjacent to a vacancy site, are bright, perfectly photostable fluorophores that exhibit high-contrast ODMR, insert of **Fig. 4c)**. The NV spin-triplet ground state features a zero-field splitting D = 2.87 GHz between states ms = 0 and ms = ±1. Laser illumination (500-650 nm) produces a spin-conserving transition to the first excited triplet state, which in turn leads to far-red photoluminescence (650-800 nm). Intersystem crossing to metastable singlet states takes place preferentially for NV centers in the ms = ±1 states, ultimately resulting in an almost-complete transfer of population to the ms = 0 state; this same mechanism allows for optical readout of the spin state via spin-dependent fluorescence [2]. The application of an external magnetic field breaks the degeneracy of the ms = ±1 state and leads to a pair of transitions whose frequencies depend on the magnetic field component along the N-V symmetry axis. There are four sub-ensembles of NV centers with different symmetry axes; thus a full ODMR spectrum contains complete information about all three vector components of the external magnetic field. My group will build a diamond magnetic microscope platform as depicted in **Fig. 4c)**. The microscope operates at room temperature and features total-internal-reflection laser side illumination (to prevent sample photodamage protection), high NA oil immersion objective (spatial resolution ~ 300 nm), scattered blue light illumination for reflectance imaging, electromagnetic coils for static and dynamic magnetic measurements, and a fast sCMOS camera for wide-field (up to 200 x 200 um^2) detection in sub-ms-second timescales. The spatial resolution of diamond magnetic imaging is limited by optical diffraction; two neighboring sources can only be resolved if they are separated by >300 nm. However isolated magnetic sources that are smaller than the diffraction limit (e.g. MNPs) can still be detected, provided they produce a sufficiently large magnetic field to be detected. We will use three modalities for mapping the magnetic field or current distribution:

1. The sample can be fabricated or deposited directly on diamond chip doped with a 50 -200 nm-thick NV sensing layer as in **Fig. 4c)** and upper of **Fig. 4d)**. We will start with pure electronic grade diamond ([N]<5 ppb) from Element 6, and implant $^{15}N^{+}$ ions at several different energies at Cutting Edge Inc. This will create a uniform surface layer of ^{15}N and lattice vacancies [32]. After the implantation we will anneal the samples at 800-1100 $^{\mathbf{o}}$C to promote NV creation while minimizing formation of parasitic paramagnetic spins.
2. In the second modality a diamond nanostructure (nanobeam, nanoparticle) will be positioned on the target sample to study, such in the left of **Fig. 4d)**. Nanodiamonds can be purchased commercially or fabricated using electron beam or optical lithography and lift-off techniques.
3. The NV center can be used in a scanning-probe configuration, such described in **Fig. 4d (lower right)**, [1].

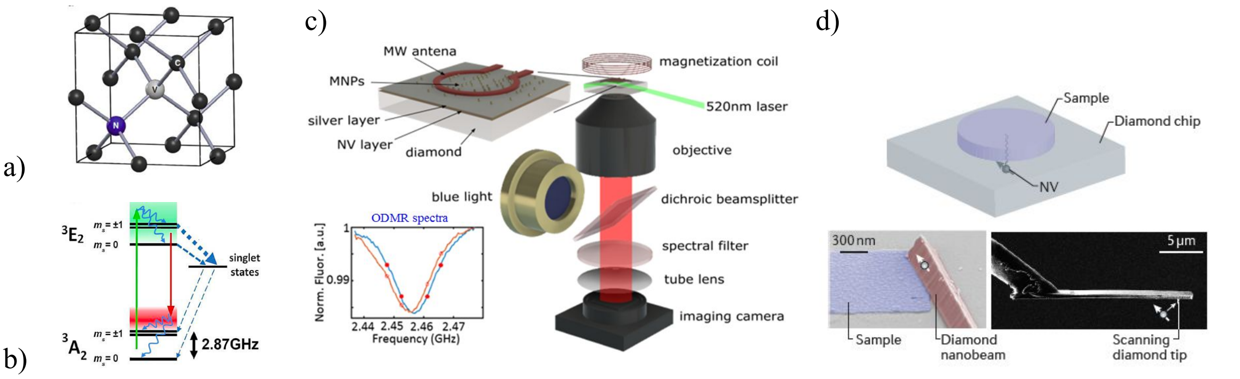


Figure 4: NV sensing schemes. (a) NV center in the diamond lattice. (b) Energy-levels of the NV center. The NV spin is pumped into the $|0\rangle$ state by off-resonance optical excitation, the $|\pm1\rangle$ excited states can decay non-radiatively through metastable singlet states, and the ground-state spin can be manipulated by microwave excitation. (c) wide-field NV magnetic microscope. Insert ODMR spectra of single NV center at a magnetic field of ~ 150 G (blue line) and in the presence of magnetic nanoparticle clusters deposited on the diamond surface (red line). d) **measuring schemes**: NV centers can be brought within a few nanometres of the sample using different approaches: the sample can be fabricated directly on diamond, a diamond nanostructure can be positioned on the sample or NV centers can be used in a scanning-probe configuration.

II.b Probing static and dynamic magnetic textures

Determining the static spin configuration of a magnetic system is a central problem to the nanomagnetism community and is crucial for the development of magnetic devices. Powerful techniques for real-space probing of nanoscale magnetic textures include MFM, XMCD and STM. NV magnetometry provides an alternative approach that is magnetically non-perturbative and works under a wide range of magnetic fields and temperatures. My group will explore novel interesting magnetic systems including: **skyrmion states in ferromagnetic thin films**, **spin structures of multiferroic thin films** (BiFeO3, FeGaB, etc.).

(a) Magnetic skyrmions are nanoscale spin textures characterized by a topological number that is invariant under continuous deformations [21]. A skyrmion creates a region of reversed magnetization in an otherwise uniform magnet and is characterized by a helicity and a chirality, similar to domain walls. Skyrmions can occur as ground states of 2D magnetic systems in the presence of chiral magnetic interactions such as the

Dzyaloshinskii–Moriya interaction, **Fig. 3a)**. They are stable near room temperature, and could possibly be memory storage units in future information technology nodes. Incorporation of magnetic skyrmions instead of magnetic domain walls may improve device performance and scaling possibilities due to their insensitivity to defects pinning. However, determining the spin texture of technologically interesting skyrmions in thin magnetic films is challenging owing to the need for a resolution of ~10–100 nm and magnetic field compatibility of the probing technique.

(b) Multiferroic films are heterostructures in which antiferromagnetism coexists with ferroelectricity, enabling an efficient electrical control of magnetization through magnetoelectric coupling [33]. Multiferroic materials are currently emerging as a unique platform for information processing, spintronic and magnonic devices. The ferroelectric properties of these materials have been widely investigated by piezoresponse force microscopy, revealing unique domain structures and domain wall functionalities [34], however the corresponding nanoscale magnetic textures and their potential for spin-based technology still remain hidden. My group will use NV magnetometer to probe the nanoscale spin-texture of multiferroics such BFO and FeGaB.

II.c Probing current distribution

In mesoscopic condensed matter systems, the spatial distribution of electrical currents plays a prominent role in some of the most intriguing known physics phenomena. My group will explore edge currents in quantum Hall systems and surface spin currents generated in topological insulators [23] such in **Fig. 3c (lower right)**. The strong spin−orbit coupling in three-dimensional (3D) topological insulators (TIs) leads to insulating bulk and conducting surface states protected by time reversal symmetry [35]. Electrons populating these topological surface states possess only one spin state per momentum state (spin-momentum locking) in contrast to conventional materials [36], which make them robust against most perturbations from defects or impurities and can enable the propagation of dissipationless spin currents. The direct electrical detection of a current-induced spin polarization in 2D and 3D TIs has been so far restricted to cryogenic temperatures [37] which limits further progress in this research field and its application potentials. **The room temperature detection** of such highly correlated spin systems is not only interesting for fundamental research but also for applications in quantum spintronic devices. My group will use both magnetoresistance and NV measurements techniques. For the later technique, the 2D current distributions can be reconstructed from the measurement of any component of the stray field in a plane above the sample. We will use the inversion procedures [38] to reconstruct the current flow in these materials and map electrons transport at the nanoscale.

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