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Materials and devices for integrated room temperature quantum spintronics

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Abstract: Recent advances in precise stoichiometry control and high-resolution characterization of advanced spintronic materials allowed for the development of integrated spintronic devices, which might enable ultralow power magnonic devices with multi-THz spin wave bandwidth and topologically protected spin wavefunctions that are robust for fabrication imperfections. In addition, advances in microwave and optical excitation and control of quantum states in diamond nitrogen-vacancy systems (diamond-NV) allowed for ultrasensitive magnetometry and integrated quantum logic applications. Here, an integrated spintronic garnet/diamond-NV quantum system has been reviewed and discussed for logic and memory applications. After an overview of the recent advances in the growth and characterization of insulating magnetic iron garnets, previous computational demonstrations of ultrawide bandwidth topologically protected few-nanometer size chiral spin structures called skyrmions are discussed for carrying information on chip between diamond-NV systems. Next, earlier diamond-NV characterization studies using microwave ferromagnetic resonance and photoluminescence measurements were reviewed. Finally, a brief discussion is presented on the steps needed for integrated quantum spintronic devices to operate at room temperature.

Key words: Quantum systems, spintronics, magnonics, diamond nitrogen-vacancy centers

1. Introduction

Practical physical implementations of quantum systems could help achieve unprecedented quantum computational advantage, ultrahigh sensitivity as well as secure communication on chip [1,2]. Many different routes have been explored for implementing quantum systems using photons, atoms, spins, mesoscopic superconducting, and nanomechanical structures [3]. All of these systems except for the photonic fiber-optical networks or free-space optical systems could be operated only at cryogenic temperatures and require significant excitation and measurement infrastructure in quantum entanglement and coherence measurements [1,3]. This large barrier for quantum system implementation prevents both the investigations of the new physics of quantum systems and the scalable development of practical quantum computers and sensors. As a result, new schemes for implementing room temperature quantum systems are needed.

Materials that host quantum emitters or discrete quantum states are essential testbeds for quantum phenomena. These materials include diamond-based doped crystals, indium arsenide quantum dots, photonic cavities, nuclear spins, superconducting tunnel junctions (Josephson junctions). The quantum phase of the states stored in these systems must be controlled on demand via strong quantum correlations with other states or external driving circuits such as microwave resonators, external magnetic field control (in nuclear magnetic resonance), and photonic cavities and polaritons in nanoelectromechanical resonators.

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Common implementations of quantum systems include hybrid architectures of quantum materials, in which qubits are stored in discrete states and a medium carries information for spin-spin or spin-photon quantum correlations or entanglement. Figure 1 shows some of the main hybrid quantum systems, which include suspended optical nanocavities and fiber optical networks, suspended optical nanocavities and Rydberg state transitions, trapped ions, superconducting quantum interference devices coupled with microwave LC resonators, quantum dot-based electronic spin ensembles coupled with microwave oscillators, and nuclear spins. These systems together span more than 12 orders of magnitude in excitation frequencies from kHz to above PHz ($>10^{15}$ Hz) as well as coherence times spanning picoseconds to few seconds. The dynamic control of the resonators in the coupled hybrid quantum systems help tune the phase of the discrete qubit states stored in the quantum media. Thus, fundamental operations such as initialization, controlled NOT, and writing operations can be established.

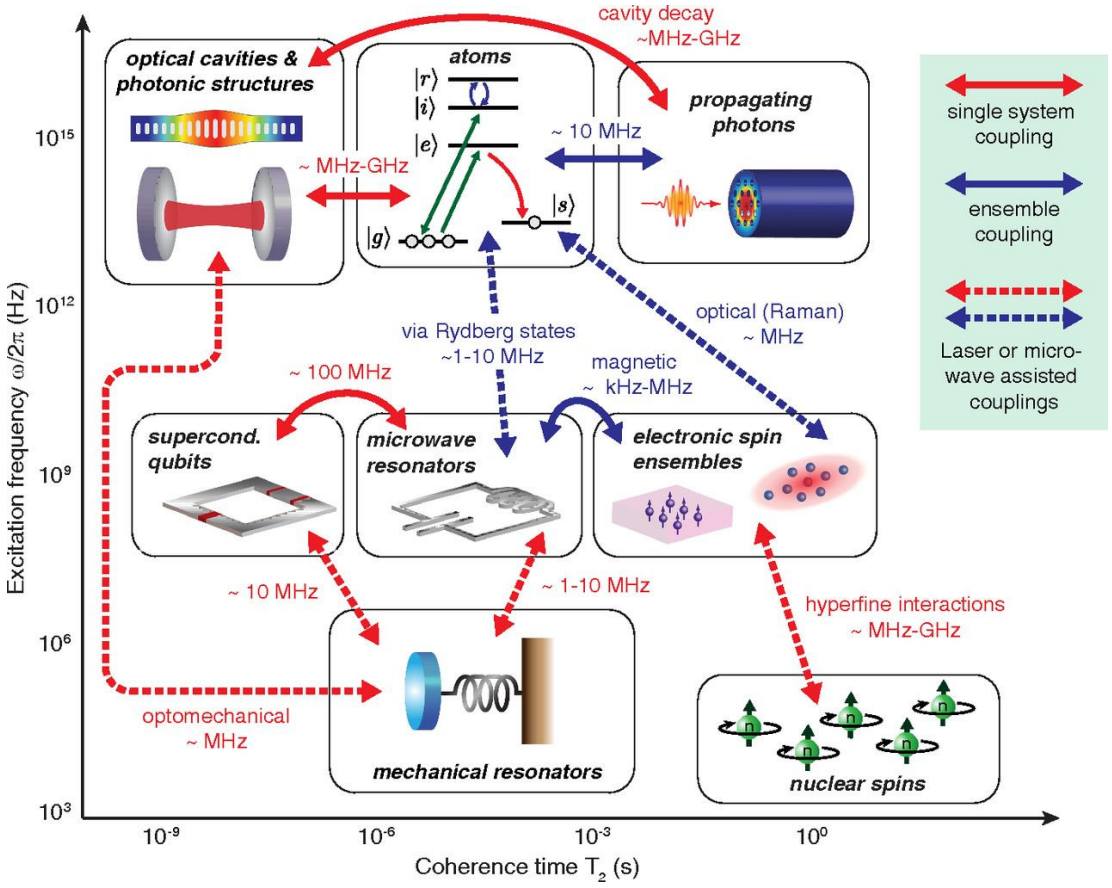

Figure 1. Quantum system implementations rely on hybrid coupling of media, which carry and store qubit states. These implementations span a vast window of excitation frequencies (kHz to optical frequencies) and coherence times (nanoseconds to more than a second). Despite their unique physics, most of these implementations only work at cryogenic temperatures, which limit both the fundamental investigations of quantum phenomena and practical quantum logic device applications. Reproduced with permission. Copyright 2015, National Academy of Sciences [3].

While a wide variety of quantum phenomena and circuits could be tested using these hybrid quantum systems, these systems require few Kelvin temperatures, high external magnetic fields, and vacuum conditions to operate with extensive optical, microwave, and magnetic driving circuits and infrastructure. On the other hand,

diamond-NV systems allow for arbitrary positioning of NV center locations via ion implantation, long quantum coherence times (up to more than one second), optical excitation (530 nm), photoluminescence detection (633 nm), microwave readout (S_{21} transmission), and room temperature spin-spin quantum correlations and entanglement. Chip-scale integration of room temperature quantum systems based on spin-spin correlations could be feasible in diamond-NV systems. Using the recent advances in spintronic materials and the topological protection in their propagating spin structures, integrated spintronic garnet/diamond-NV systems have been discussed as promising platforms in this review for room temperature quantum logic and memory applications.

In the next section, an overview is provided on the properties of the candidate quantum materials and spintronic materials which could make up the qubit medium and a bath medium for carrying magnons, respectively. In Section 3, the coupled diamond-NV system and the spintronic bus line as a promising platform is presented. In Section 4, the previous experimental studies on the coupled diamond-NV systems are presented. Finally, the necessary steps for integrated quantum spintronic devices to operate at room temperature are discussed as the outlook.

2. Proposed system architecture

Quantum systems need to embody the electronic or nuclear spin qubits in desired positions on chip, which can be determined by nitrogen ion implantation in diamond and annealing [4]. In a proposed architecture, a bilayer of shallow diamond-NV centers and a magnetic insulator iron garnet thin film make up a structure for qubit initialization, storage, spin wave-based entanglement, and readout as shown in Figure 2. The magnetic film must be an insulator with ultralow conductivity, wide electronic bandgap and out-of-plane magnetic easy axis for carrying chiral spin waves called skyrmions [5,6]. In diamond, the NV centers could be spaced either in very close proximity to function as an ensemble qubit or they could be more than a few micrometers apart. In the sparse NV distribution, the spins are coupled mainly by dipolar interactions among each other. Skyrmions in the magnetic film under diamond-NV system can interact via dipolar interactions. In the following two sections, earlier experimental results on the structure mentioned are presented and discussed.

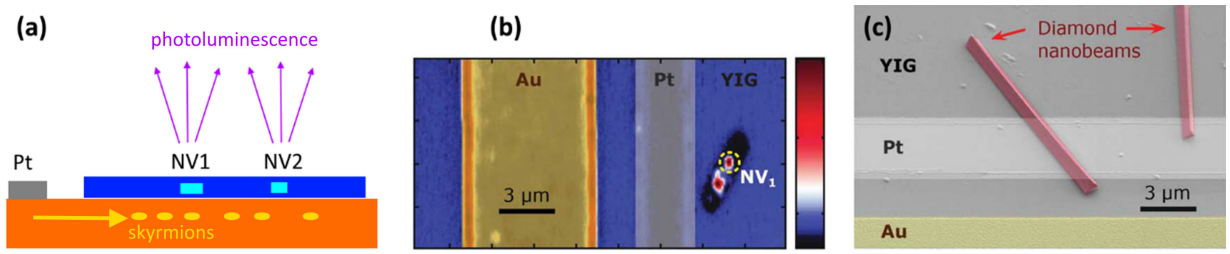

Figure 2. Proposed architecture for skyrmion-mediated quantum correlations and entanglement between diamond-NV centers. (a) Pt layer or a 2D layer with large spin-orbit coupling excites the skyrmions. The dipolar coupling between the skyrmions along the garnet/diamond interface and the NV electronic spins can drive the orientation of the NV electronic spins. NV centers are coupled via the spin waves and skyrmions and their dipolar terms in the Hamiltonian for optimized NV distances, microwave drive power, and pulse width as well as magnetic layer properties (saturation moment and skyrmion pulse profile). (b) Photoluminescence map of NV centers [7]. (c) False-colored scanning electron micrographs of diamond-NV nanobeams on garnet layer. Reproduced with permission. Copyright 2017, American Association for the Advancement of Science [7].

3. Available materials for quantum computation

Quantum correlations among NV centers can be established within the same diamond crystal using dipolar interactions and strong proximity effects such as exchange coupling without any mediation from a different spintronic material [8]. While these systems were demonstrated already for strong quantum correlations, these NV systems must be used in conjunction with a materials platform for microwave spin wave addressing (write) and readout. Thus, the available materials for room temperature quantum computation are investigated in two parts: quantum materials for storing qubits and spintronic materials for addressing/reading out the electronic spin states in these quantum materials.

3.1. Quantum materials

Quantum materials are defined as the hosts for discrete quantum states and emitters, such as nitrogen-vacancy, silicon-vacancy, tin-vacancy and lead-vacancy centers in diamond, InAs quantum dots and others [1,9]. For quantum computation purposes, the host must have a wide electronic bandgap, very low phonon density of states and large Debye temperature to achieve long coherence times, while the spin phase in the qubit materials should be accessible via optical and microwave excitation and readout. As a result, diamond and group-IV dopants with vacancy centers [10], InAs quantum dots, quantum dots in transition metal dichalcogenides, such as WSe_2 [11] and optically active and spin-active defects in SiC [12] emerge as main candidates for room temperature spintronic quantum circuits. SiC processing and reproducible sample preparation poses additional challenges than other materials platforms; therefore, SiC has not been included in the candidate quantum materials list.

Diamond and and group-IV dopants with vacancy centers [10]: Diamond has been shown to achieve room temperature entanglement between nitrogen and vacancy centers [8]. These materials contain a nitrogen substituting a carbon atom and a vacancy of carbon atom adjacent to the nitrogen atom. The complex is negatively charged (NV^{--}).

Table 1 shows a summary of candidate material systems and their properties [1]. These properties include spin coherence time, which is the time over which spins retain a predictable phase (i.e. over Rabi oscillations) before undergoing scattering. Spin coherence is an important property in establishing cascadable quantum logic gates and circuits. Ancilla qubits are the reference bits whose states are previously known and are never changed; thus, the state of the spin qubits which store or process information are always compared to the ancilla qubits. These ancilla qubits must be stable for storing entangled states or quantum error correction. Measured linewidths are the full-width at half-maximum across the emission resonance peaks for each of the quantum emitter/bit. Quantum emission peaks at the energies with zero phonon density are called ZPL emission (ZPL: zero phonon line). These states do not scatter with phonons as they have no overlap with phonon states at those wavelengths. ZPL emission wavelength is the wavelength at which photons are emitted due to spin relaxation and is a function of the zero-field splitting.

An important material group not included in this table is hexagonal boron nitride (hBN) monolayers with defects (nitrogen vacancy or antisite nitrogen vacancy), which are also promising material platforms due to their wide band gap (6 eV), which allow for subbandgap green laser excitation (532 nm) to avoid excitonic emissions and achieve bright room temperature zero phonon line emission at 623 nm due to defects. These monolayer materials could allow for direct access to the spin states via exchange bias or near-field dipolar interactions from the spin waves from spintronic underlayers.

Table 1. Candidate platforms for room temperature integrated spintronic quantum systems, their coherence times, microwave resonance, and emission characteristics (reproduced from ref. [1].)

Material	Spin coherence time (T_2)	Ancilla qubits	Measured linewidth (MHz)	Emission fraction into ZPL	ZPL emission waveleng. (nm)	Radiative quantum efficiency
Diamond-NV	>1 s	^{13}C ve ^{14}N	14	∼0.03	637	>0.8
Diamond-SiV	>100 ms	^{29}Si ve ^{13}C	∼100	>0.7	737	~ 0.1
Diamond-GeV	>100 ns	^{29}Ge ve ^{13}C	∼42	∼0.7	602	>0.5
SiC DV	~ 1 ms	^{29}Si ve ^{13}C	>80	∼0.07	∼1100	(N/A)
SiC V_{Si}	~ 20 ms	^{29}Si ve ^{13}C	(N/A)	∼0.4	∼900	(N/A)
Quantum dots in TMDC	(N/A)	(N/A)	∼2000	(N/A)	Near-infrared	(N/A)
InAs/GaAs quantum dots	3–10 µs	Mn dopings, quantum dots	∼300	>0.95	900-1550	∼1

Diamond-NV centers have lower emission fraction with respect to other group-IV defects in diamond or hBN, but their longer spin coherence times make this material system stand out. Therefore, this review focuses on diamond-NV systems for their narrow linewidths, long spin coherence times, their wide electronic band gaps and zero phonon emission lines in visible.

3.2. Spintronic materials

Spintronic materials are magnetic films that carry spin waves. To match the electron spin resonances in diamond-NV systems within 1-2.5 GHz, ferromagnetic or ferrimagnetic thin films are used [7]. These films are characterized by a magnetic order which minimizes its energy on the directions along the easy axis. This order is called magnetic anisotropy, and it is represented as a sum of the following terms [6]:

$$\mathcal{E}=\left[\mathrm{A_{ex}}(\nabla\mathbf{m}\left(\mathbf{r}\right))^{2}-\mu_{0}\mathrm{M_{s}}\mathbf{m}\left(\mathbf{r}\right).\mathbf{H}+\mathrm{K_{u}}\left(1-\left(\mathbf{m}\left(\mathbf{r}\right).\mathbf{e_{z}}\right)^{2}\right)+\mathrm{D_{i}}\left(\mathrm{m_{z}}\nabla.\mathbf{m}\left(\mathbf{r}\right)-\left(\mathbf{m}\left(\mathbf{r}\right).\nabla\right)\mathrm{m_{z}}\right)+D_{b}\boldsymbol{m}\left(\boldsymbol{r}\right).\left(\nabla\times\boldsymbol{m}\left(\boldsymbol{r}\right)\right)\right]$$

where m(r) is the local normalized magnetic moment orientation $\mathbf{m}\left(\mathbf{r}\right)=\frac{\mathbf{M}}{\mathbf{M_s}}$ with normalized magnitude $\left|\mathbf{m}\left(\mathbf{r}\right)\right|=1$, $\mathrm{A_{ex}}$ is the exchange stiffness coefficient, $\mathbf{H}$ is the applied magnetic field, μ_0 is the vacuum magnetic permeability, M_s is the saturation magnetization, K_u is the uniaxial anisotropy constant, $\boldsymbol{e_z}$ is the anisotropy easy axis direction. D_i and D_b are the interface and bulk Dzyaloshinskii–Moriya interaction energy densities, resulting in Neél-type (hedgehog) and Bloch-type (vortex) skyrmions, respectively. Skyrmions are chiral spin waves with topological order. Unlike spin waves, skyrmions can be stabilized along interfaces using current pulses. The first term in this energy expression is exchange interaction of adjacent spins and exchange interaction drives the propagation of spin waves in most cases. The second term is the Zeeman energy due to the applied external magnetic field. The third term is the uniaxial anisotropy term which may originate due to the growth-induced lattice strains inside the magnetic films. The last two terms are the interface and bulk Dzyaloshinskii–Moriya interaction energies, whose balance with exchange and uniaxial energies give rise to skyrmions. Examples of skyrmions are shown in Figure 3(a) on the right hand side. In this computational example, spin-polarized charge currents have been used to nucleate, grow, and laterally translate skyrmions between 114 MHz to 21 GHz. The operation frequencies could be tuned using the DC driving current intensity.

For the purposes of strong quantum correlations and long coherence between spin waves in the magnetic film and the diamond-NV center electronic spins, magnetic insulators such as garnets must be used. In addition, due to the

propagating and dispersive nature of the spin waves (magnons), spin waves such as skyrmions that have topological order should be used. Skyrmions could in principle be stabilized under diamond-NV centers on demand using current pulses.

Perpendicular magnetic anisotropy in garnet layers is needed for stabilizing skyrmions. $Tm_3Fe_5O_{12}$ films as shown in Figure 3 (b)-(d) and other garnets [13,14] allow for perpendicular magnetic anisotropy and skyrmions at room temperature. Since skyrmions can be driven in microwave frequencies such as 1-5 GHz, one can achieve electron spin resonance coupling of the skyrmion modes. Thus, electron spin resonance splitting can be established based on the presence of skyrmions, and these modes might help control the phase of the NV electronic spins.

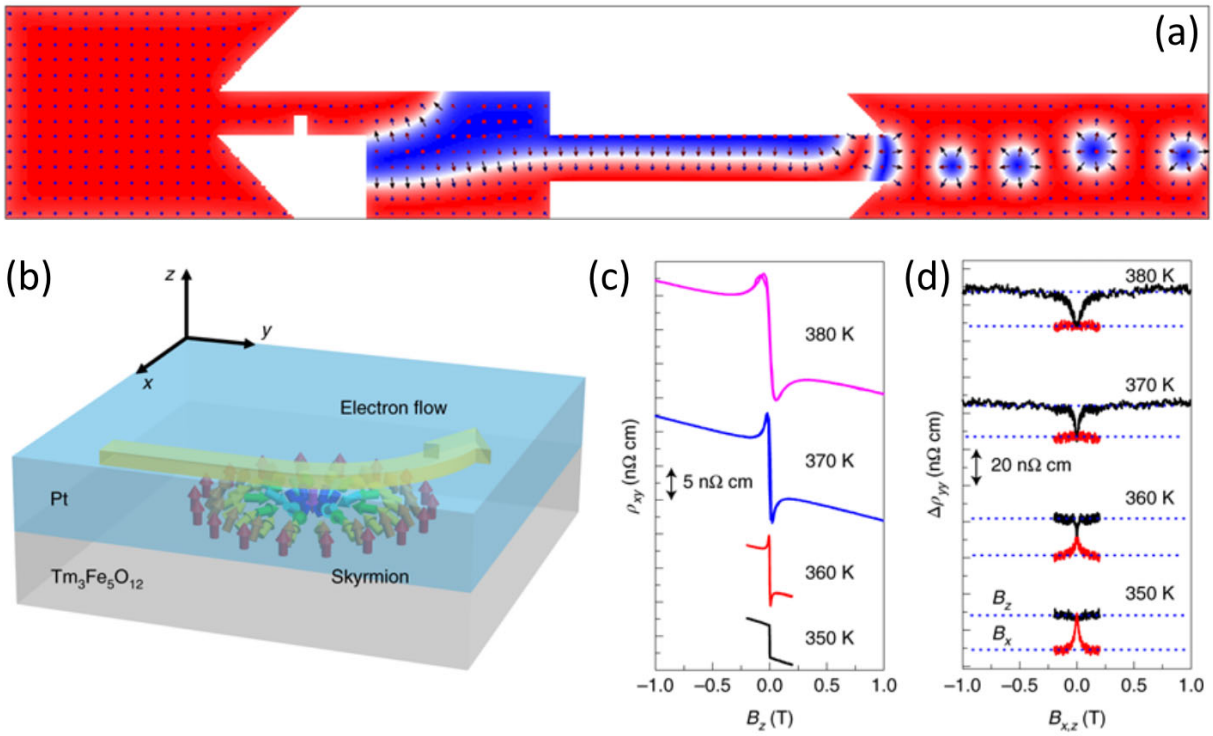

Figure 3. Skyrmions along interfaces as drivers for quantum correlations. (a) Skyrmions can be driven along Co/Pt interfaces using spin-polarized DC current. (b) The layers in part a must be substituted with a magnetic insulator such as thulium iron garnet ($Tm_3Fe_5O_{12}$) which can also stabilize skyrmions along its interface with Pt. Spin Hall resistivities of $Tm_3Fe_5O_{12}$/Pt shows strong switching characteristics under external (c) vertical and in-plane (d) transverse and longitudinal magnetic fields. Reproduced with permission. Copyright 2019, Springer Nature [5].

4. Diamond film characterization

Previous studies [7] probed the magnetic-noise spectrum of a magnetic garnet film ($Y_3Fe_5O_{12}$, in-plane easy axis) without external drive fields. The NV spin relaxation rates were measured as a function of an external magnetic field B_{ext} along the NV axis, which are shown in Figure 4 (a). Figure 4 (b) explains the transitions shown in Figure 4 (a). The peak in the relaxation rate for NV1's $m_s = 0 \leftrightarrow -1$ transition (m_s: electron quantum number) is attributed to the high density of spin waves just above the ferromagnetic resonance frequency of the magnetic layer when the corresponding electron spin resonance (ESR) frequency crosses this region (Figure 4 (b)). Figure 4 (c) shows the normalized photoluminescence of the diamond-NV/garnet bilayer system under external applied magnetic field and the applied microwave drive frequency on the garnet film. The photoluminescence plot shows the ESR of NV1 and the ferromagnetic resonances of the garnet film. Further characterization of a similar bilayer of diamond-NV/garnet with

perpendicular magnetic anisotropy is needed under optical and magnetic initialization of the NV electronic states and the microwave excitation of skyrmions in the garnet layer would help elucidate the quantum correlations among NV1, NV2, and the skyrmions.

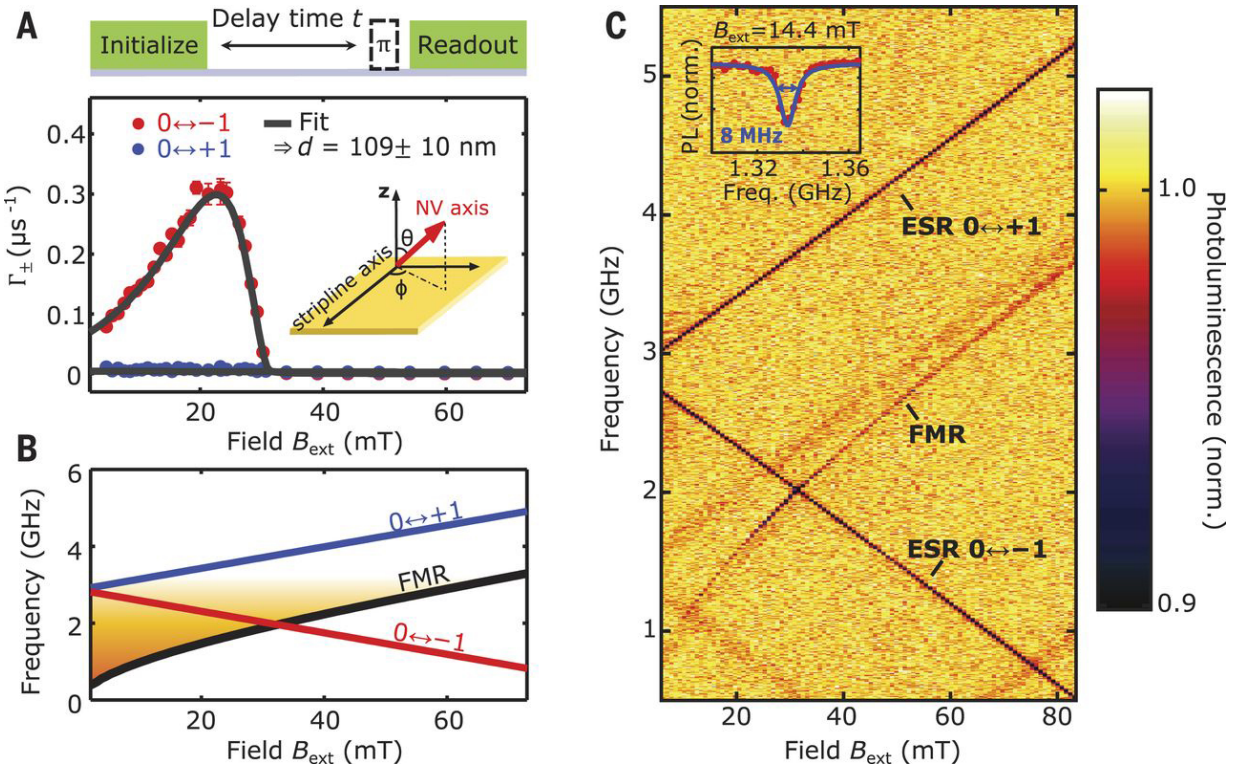

Figure 4. Spin waves along interfaces as drivers for quantum correlations. (a) Measured spin relaxation rates for an NV center for $m_s = 0 \leftrightarrow -1$ and $m_s = 0 \leftrightarrow +1$ transitions. (b) Magnon density as a function of frequency and applied vertical external magnetic field together with the electron spin resonance (ESR) transitions ($m_s = 0 \leftrightarrow \pm 1$). (c) Normalized photoluminescence (PL) of NV1 as a function of external magnetic field and the frequency of an applied microwave drive field on the garnet layer. The labeled straight lines correspond to the NV ESR transitions in the electronic ground states. The inset shows the 8 MHz linewidth of the YIG FMR under $B_{ext} = 14.4$ mT. Reproduced with permission. Copyright 2017, American Association for the Advancement of Science [7].

5. Conclusion and outlook

This review overviews the candidate quantum materials for room temperature integrated quantum computation. The fundamental discrete quantum nature, superposition, and entangled states need to be established at room temperature despite significant decoherence channels. Magnetic insulator/diamond-NV systems emerge as an important and promising candidate platform for room temperature quantum computation. Earlier studies showed that magnetic insulator iron garnets with perpendicular or in-plane magnetic easy axis can be stabilized at room temperature. A bilayer of diamond-NV system and a magnetic insulator underlayer could facilitate quantum correlations between the NV centers based on their relative positions, microwave driving, external magnetic field, and the magnetic film properties (saturation moment, exchange coefficient). Diamond-NV systems provide the perfect insulating platform with long coherence times for room temperature electronic and room temperature spin qubit states while skyrmions could help achieve microwave-based driving, topological protection, readout and coupling on demand. In order to establish room temperature quantum correlations among the NV centers and the skyrmions, coherent coupling between the NV center electronic spins and

the skyrmions must be demonstrated with excellent NV and diamond quality, strong interface DMI effects, as well as magnetic insulator materials with large room temperature magnetic anisotropy.

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