Ajoy's research program has focused on improving the spatial resolution and sensitivity of NMR via coupling to optical degrees of freedom, hyperpolarization, and the excellent coherence properties of nuclear spins in unusual regimes. Some of these are highlighted below:

"Quantum sensor" magnetic resonance: Ajoy's early work focused on fundamental research developing quantum sensing methodologies with the aim of shrinking down the sensitive length scale of NMR from macroscale radio-coils to nanoscale volumes. This leveraged developments in the quantum information field, where Nitrogen Vacancy (NV) defect centers in diamond—highly coherent *single* electron spins whose spin state can be initialized and read out optically—were developed as qubit candidates. Experiments were centered on NV-center mediated optical detection of magnetic resonance signals at sub-micron length scales [1].

Ajoy made several impactful contributions in choreographing spin dynamics in these quantum sensor systems to efficiently extract information of practical relevance, including novel methods for "Hamiltonian tomography," towards unraveling the couplings between spins in a network, using relayed polarization walks through the network [4]. Another noteworthy method was "*quantum interpolation*" [5], which enabled linear interpolation of the Hamiltonian dynamics of a quantum sensor to boost the frequency (and hence spatial) resolution of nano-NMR. Ajoy proposed the ability of optically polarized ^{14}N nuclear spins in diamond to serve as rotational sensors (gyroscopes) [7-9], with advantages of high stability, optical interrogation, and ability to interface with existing MEMS devices.

Room temperature optical hyperpolarization in nanoparticle media: Ajoy has made noteworthy contributions to optical hyperpolarization methodologies. This exploits the ability of optically polarizable electrons (e..g NV centers) to be *completely* (100%) optically polarized at room temperature, even at Earth's magnetic field. Transferring this polarization to nuclei of interest can hyperpolarize them into athermal spin populations with vastly enhanced NMR signals. Ajoy contributed to practical and theoretical approaches for "orientation independent" hyperpolarization from optically polarizable electrons [10], demonstrating hyperpolarization of diamond powder. This employed multiple frequency sweeps at low field [11], was remarkably robust and simple to produce, and introduced new mechanistic features to dynamic nuclear polarization [12] because it was carried out in a low-field regime where the electron-nuclear hyperfine coupling is of the same order of magnitude of the nuclear Larmor frequency. This produced a miniature solid-state device to generate this hyperpolarization in NV-rich nanodiamonds [13]. Hyperpolarization also unraveled fundamental aspects of the mechanisms driving nuclear relaxation in electron-rich solids [14], with broad implications.

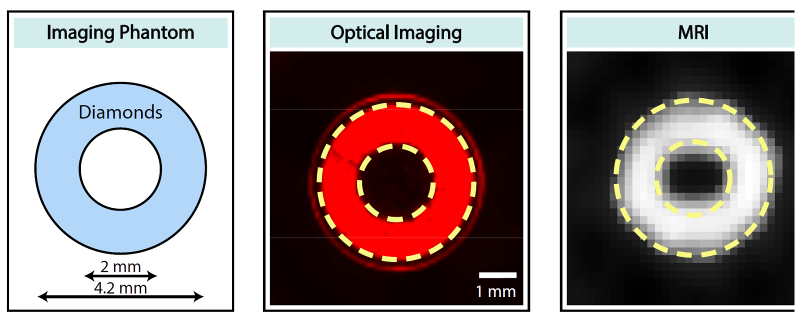

Figure 1: Dual-mode optical and MRI imaging. (A) Ring-shaped phantom filled with 40mg diamond particles (B) *Fluorescence image* captured through a 630nm long-pass filter. (C) **^{13}C** MRI image.

Hyperpolarized nuclear spin sensors and imaging agents: Subsequent work in Ajoy's independent research group at UC Berkeley has further explored the fundamentals and technological applications of optical hyperpolarization. One recent research direction combined optical and magnetic resonance (MR) imaging in a "dual" imaging mode [15] **(Fig. 1)**. These are attractive in combination because they offer complementary advantages of resolution and speed, especially in the context of imaging in scattering environments. The experiments employed microdiamond particles that fluoresce brightly under optical excitation while ^{13}C is simultaneously hyperpolarized, making them bright under MR imaging. Experiments demonstrated the inherent advantages of the dual-mode approach to allow background-free particle imaging and showed that, because the two imaging modes proceed in Fourier-reciprocal domains (real and k-space), the combination of the two modes can accelerate image reconstruction in sparse-imaging scenarios.

Another effort recently showed that, once hyperpolarized, strongly interacting nuclear spins attain long coherence times, which can be exploited for practical purposes. Ref. [16] reported the observation of long-lived transverse spin states in hyperpolarized ^{13}C nuclei, showing extremely long lifetimes T_2'>90s at room temperature [16]. These experiments involved quasi-continuous observation of the ^{13}C nuclei along with application of >6M

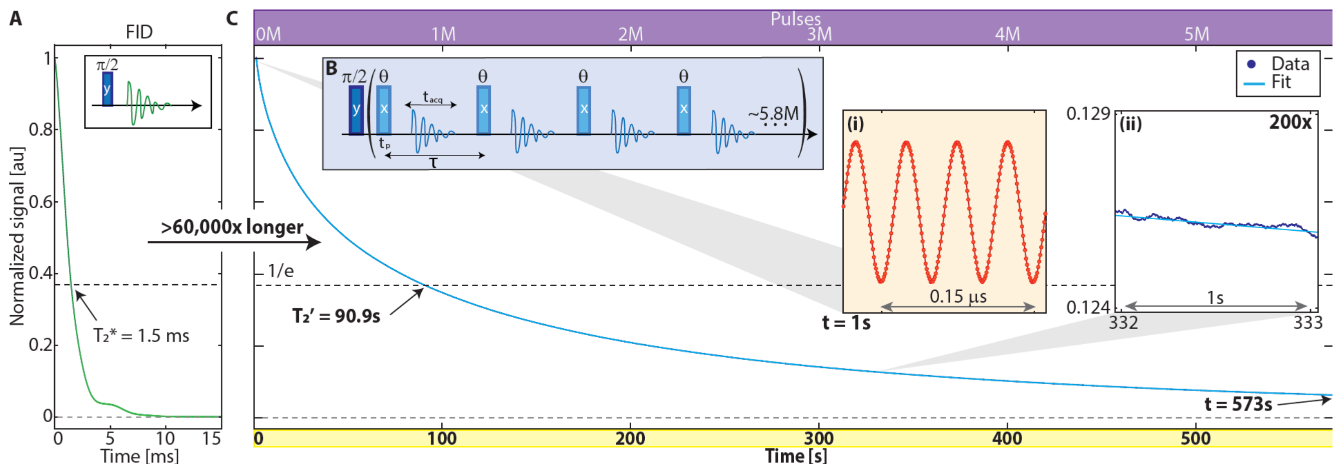

Figure 2: Ultralong ^{13}C transverse lifetimes. (A) Conventional ^{13}C free induction decay with T_2^*~1.5ms. (B) Floquet drive consists of a train of pulses applied spin-locked with the ^{13}C nuclei. Spins are interrogated in windows between the pulses, with the nuclear precession sampled every 1ns. (D) *Minutes-long lifetimes* of the transverse state. Data (blue points) shows *single-shot* measurement and solid line is a fit. Here we apply ~6M pulses (upper axis). *Inset (i):* Raw data showing measurement of the ^{13}C spin precession, here at 1s into the decay. *Inset (ii):* Data zoomed 100x in a 1s window demonstrates high measurement SNR. Using a 1/*e* -intersection as a proxy for the decay lifetime, we estimate T_2'~90.9s

pulses and demonstrated a lifetime extension over the free induction decay by >60,000-fold **(Fig. 2)**. The combination of minute-long transverse nuclear lifetimes and continuous interrogation engendered new quantum sensors, [17] where densely packed, hyperpolarized ^{13}C nuclei are exploited as sensitive magnetometers of time-varying fields in high bias-field environments (1-20T). Newer work has revealed interesting features in these sensors that exploit many-body dynamics between the dipolar-coupled hyperpolarized nuclei.

[1] Ajoy et al., *Phys. Rev. X* **5**, 011001 (2015); [2] Lovchinsky et al., *Science* **351**, 836 (2016); [3] Abobeih et al., *Nature* **576**, 411 (2019); [4] Burgarth and Ajoy, *Phys. Rev. Lett.* **119**, 030402 (2017); [5] Ajoy et al., *PNAS* **114**, 2149 (2017); [6] Glenn et al., *Nature* **555**, 351 (2018); [7] Jaskula et al., *Phys. Rev. Applied* **11**, 054010 (2019); [8] Ajoy and Cappellaro, *Phys. Rev. A* **86**, 062104 (2012); [9] Jarmola et al., *Sci. Adv.* **7**, eabl3840 (2021); [10] Ajoy et al., *Sci. Adv.* **4**, eaar5492 (2018); [11] Ajoy et al., *PNAS* **115**, 10576 (2018); [12] Zangara et al., *PNAS* **116**, 2512 (2019); [13] Ajoy et al., *Rev. Sci. Instr.* **91**, 023106 (2020); [14] Ajoy et al., *Nature Comm.* **10**, 1 (2019); [15] Lv et al., *PNAS* **118**, e2023579118 (2021); [16] Beatrez et al., *Phys. Rev. Lett.* **127**, 170603 (2021); [17] Sahin et al., *Nat. Comm.* 13, 5486 (2022)