Optically Detected Magnetic Resonance

Quantum spin probe of single charge dynamics

Jonathan Marcks, David Awschalom (University of Chicago)
Mickey Martini, Igor Shavrin (Swabian Instruments GmbH)

Dopants in semiconductors play a key role in modern technology and quantum applications. Many of these dopants carry spin momentum, and when materials like diamond are doped with deep-level defects, it allows for precise manipulation of spin states in optically active defects. These color centers can interact with nearby dark spins, which may induce decoherence but also offer opportunities to enhance quantum sensing and memory technologies. To explore these interactions, optically detected magnetic resonance (ODMR) is a powerful experimental technique, offering precise control and detection of quantum states in systems like nitrogen-vacancy (NV) centers in diamond. ODMR measurements require highly precise and flexible generation of microwave and optical pulse sequences, especially when complex timing and synchronization are necessary. The *Pulse Streamer 8/2* and the *Time Tagger*s by *Swabian Instruments* are designed to meet the demanding requirements of ODMR experiments [1-6].

This Application Note is a collaborative effort to showcase how Awschalom Group at the University of Chicago has leveraged *Swabian Instruments*' *Pulse Streamer 8/2* to probe the dynamics of single charges in diamond with nearby NV centers at room temperature [1]. By delivering the exact pulse sequences necessary for time-resolved double electron-electron resonance (DEER) and Hartmann-Hahn resonance measurements, the *Pulse Streamer 8/2* enables the identification and disentanglement of complex interactions in quantum systems with single particle resolution.

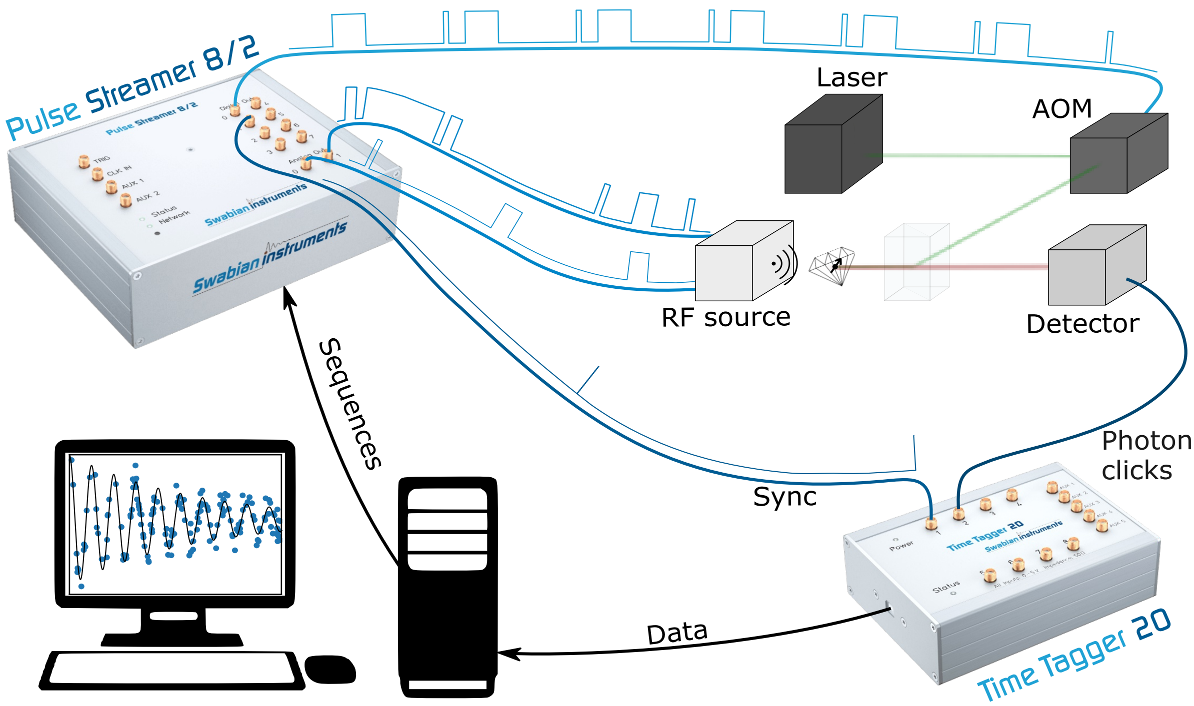

Fig. 1: Illustration of an experimental setup for pulsed Optically Detected Magnetic Resonance (ODMR). The setup consists of a laser excitation source modulated by an acousto-optic modulator (AOM) and synchronized with radio-frequency (RF) pulses, which are analog-modulated and delivered via an antenna. Photoluminescence from the sample is detected by either a single-photon detector or a photodiode, depending on the signal intensity. The Pulse Streamer 8/2 provides precise timing, synchronization, and control of the RF pulses, enabling advanced ODMR experiments.

Experimental Setup

Figure 1 illustrates an experimental setup for ODMR measurements. The *Pulse Streamer 8/2* generates pulse patterns to modulate the radio-frequency (RF) switches and acousto-optic modulator (AOM), and to synchronize the data acquisition. With its 8 digital and 2 analog channels, the *Pulse Streamer 8/2* offers sub-50 ps timing precision, ensuring accurate synchronization between microwave pulses and laser excitations. Custom pulse patterns can be easily defined and visualized with just a few lines of code using the intuitive *Pulse Streamer* API, which is supported by many programming languages. For guidance on pulse pattern definition and generation with the *Pulse Streamer 8/2*, as well as data acquisition and analysis using *Swabian Instruments*' *Time Tagger*s with picosecond timing resolution, please refer to the ODMR tutorial, available under this link: https://www.swabianinstruments.com/static/documentation/TimeTagger/tutorials/ODMR.

Measurement and Results

To investigate the charge dynamics of single nitrogen centers (N_s) in diamond, a spin probe is developed focusing on charge state multiplicity and non-equilibrium dynamics through the resonant detection of associated spin polarization using a nearby NV color center. The NV centers and N_s centers (referred to as P1 centers in their spin-1/2 neutrally charged state) are confined in a thin nitrogen δ-doped region of isotopically purified diamond (Fig. 2a).

A pump-probe sequence with dual microwave signal generators is leveraged to achieve spin polarization of the N_s center with the NV center spin, followed by a polarization-sensitive NV coherence measurement. Figure 2b illustrates the pulse sequence protocol, including the Hartmann-Hahn sequence for polarization transfer and the subsequent DEER sequence used to probe the transferred polarization. The process begins with a Hartmann-Hahn resonant drive, where synchronized microwave pulses are applied simultaneously to both the NV and N_s spin systems to transfer polarization coherently from the optically polarized NV center to the target N_s spin. After this transfer, the N_s spin center undergoes a period of free evolution. Next, a double electron-electron resonance (DEER) measurement, using two microwave tones, is then used to map the N_s spin polarization onto the NV spin coherence. The results display a finite out-of-phase NV spin coherence signal ($S_{\pi/2}$), which occurs when polarization is successfully transferred to the N_s center. This change in NV

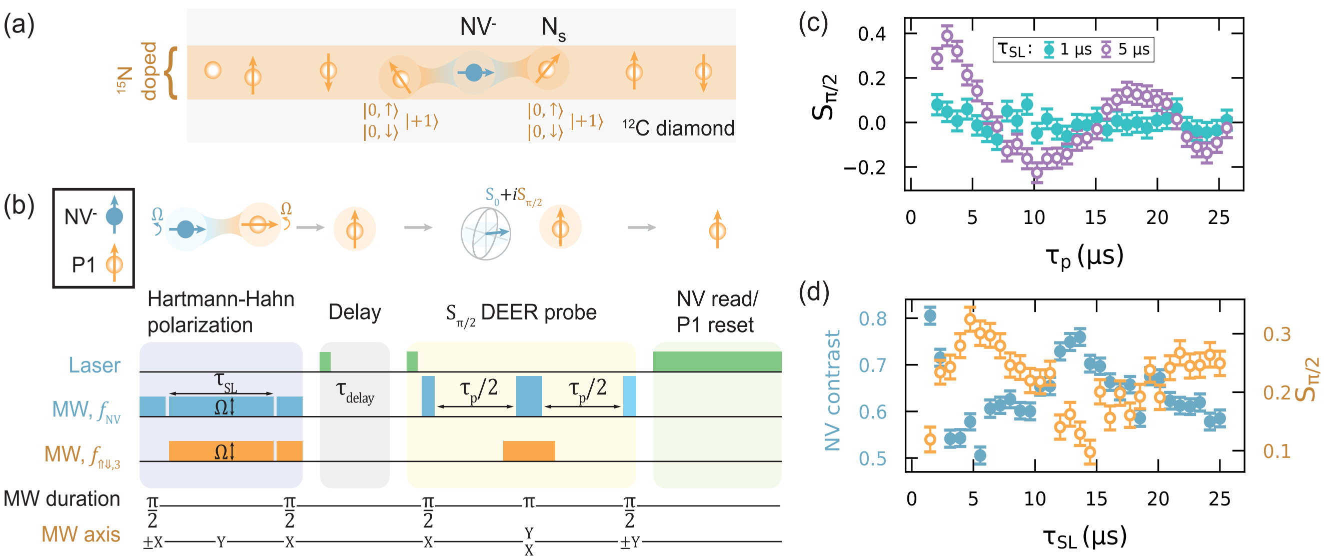

Fig. 2: (a) A nitrogen vacancy (NV) center in a N-doped layer of diamond, strongly coupled to N_s defect centers. (b) Spin pump-probe pulse sequence with interleaved laser and microwave pulses. The microwaves are IQ modulated and synchronized through the Pulse Streamer 8/2. The schematics atop the sequence demonstrate the relevant physics in the measurement. (c) The out-of-phase NV coherence, detected with ODMR, appears when polarization is transferred to the P1 center. (d) Correspondence between transferred and measured polarization signals. Figure adjusted from Ref. [1].

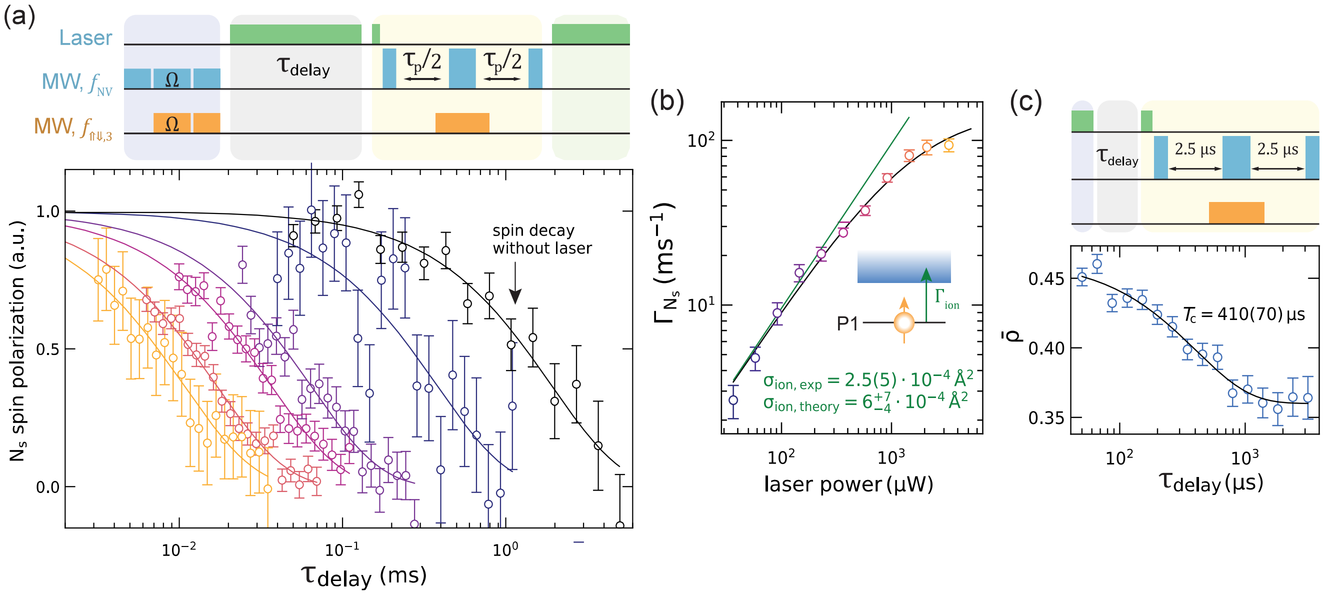

Fig. 3: (a) Spin-detected ionization decay of the Ns center measured through NV ODMR. The spin pump-probe sequence is shown on top. (b) Extracted ionization decay, with correspondence to theoretical calculations. (c) Post-ionization charge state decay measured through NV DEER. Figure

coherence is reflected in the modulation of NV fluorescence (NV contrast), indicating polarization transfer between the NV and N_s centers.

Next, this technique is applied to study non-equilibrium charge dynamics, particularly the ionization of the N_s center under illumination. The Hartmann-Hahn + DEER sequence is first employed to measure the N_s spin relaxation in the dark (Fig. 3a), revealing a T_1 (relaxation time) of approximately 2 ms, consistent with prior bulk measurements, although in this case a single spin is measured. Upon illumination with 532 nm light—gated with the *Pulse Streamer 8/2*, and energetic enough to ionize the charge—a faster relaxation of the spin polarization is observed, effectively measuring a fast process (ionization) through a steady-state spin state. By mapping the decay rate versus laser power (Fig. 3b), an ionization cross-section is extracted that is consistent with first-principles DFT calculations. Additionally, the N_s spin polarization remains long-lived under illumination, decaying only when the accompanying charge is ionized—a novel observation. Finally, charge state decay in the dark is measured (Fig. 3c), revealing a slow post-ionization decay over hundreds of μs, detected through the NV DEER signal via ODMR.

Conclusion

This Application Note demonstrates that the *Pulse Streamer 8/2* offers highly precise control and synchronization of ODMR measurements, making it a valuable tool for complex quantum experiments. The experimental results presented by Marcks, et al. at the University of Chicago not only suggest new avenues in quantum sensing and the development of qubit host materials but also highlight the potential of the *Pulse Streamer 8/2* to investigate noise sources on the diamond surface Ref. [1]. By enabling the study of coupled charge and spin dynamics—critical limitations for NV center sensing applications—the *Pulse Streamer 8/2* provides deeper insights to drive advances in NV-based quantum technologies.

References

[1] J.C. Marcks, et al. "Quantum spin probe of single charge dynamics." Phys. Rev. Lett. 133, 130802 (2024)
[2] X. Gao, et al. "Nanotube spin defects for omnidirectional magnetic field sensing." Nat. Commun. 15, 7697 (2024)
[3] J. Zhengzhi, et al. "Quantum sensing of radio-frequency signal with NV centers in SiC." Sci. Adv. 9, 2080 (2023)
[4] X. Gao, et al. "Nuclear spin polarization and control in hexagonal boron nitride." Nat. Mater. 21, 1024 (2022)
[5] G. Petrini, et al. "Nanodiamond–quantum sensors reveal temperature variation associated to hippocampal neurons firing." Adv. Sci. 9, 2202014 (2022)
[6] B. Diler, et al. "Coherent control and high-fidelity readout of chromium ions in commercial silicon carbide." Npj Quantum Inf. 6, 11 (2020)

www.swabianinstruments.com | info@swabianinstruments.com | +49 711 4004790
Swabian Instruments GmbH | Stammheimer Str. 41 | 70435 Stuttgart | Germany