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Quantum control of orbital and spin dynamics in diamond using ultrafast optical pulses[1]

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Optically addressable spin defects in solid-state materials have shown great potential for applications ranging from metrology to quantum information processing. Many of these experiments require a detailed understanding of the full Hamiltonian dynamics in order to develop precise quantum control. Here we use picosecond resonant optical pulses to investigate the coherent orbital and spin dynamics of the nitrogen-vacancy (NV) center in diamond, over timescales spanning six orders of magnitude. We implement an ultrafast optical pump-probe technique to study the NV center's orbital-doublet, spin-triplet excited state at cryogenic temperatures ($T < 20$ K), where the excited state becomes stable and optically coherent with the ground state. This technique, coupled with optical polarization selection rules, allows us to probe the coherent orbital dynamics of the NV center's excited state [1]. These experiments reveal dynamics on femtosecond to nanosecond timescales due to the interplay between the ground and excited state orbital levels. This all-optical technique also provides a method to dynamically control the spin state of the NV center by harnessing the excited state structure. Through studying the spin dynamics of the NV center with coherent pulses of light, we are able to rotate the spin state on sub-nanosecond timescales. Furthermore, by tuning the excited-state spin Hamiltonian with an external magnetic field, we demonstrate arbitrary-axis spin rotations through controlled unitary evolution of the spin state. Extending this to the full excited-state manifold, we develop a time-domain quantum tomography technique to precisely map the NV center's excited state Hamiltonian. These techniques generalize to other systems and can be a powerful tool in characterizing and controlling qubits in other optically addressable spin systems.

[1] L. C. Bassett*, F. J. Heremans*, D. J. Christle, C. G. Yale, G. Burkard, B. B. Buckley, and D. D. Awschalom, Science 345, 1333 (2014).

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