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All-optical quantum dynamical control of an NV center spin in diamond<sup>1</sup> B.B. BUCKLEY, C.G. YALE, D.J. CHRISTLE, F.J. HEREMANS, L.C. BASSETT, D.D. AWSCHALOM, Center for Spintronics and Quantum Computation, University of California, Santa Barbara, California 93106, G. BURKARD, Department of Physics, University of Konstanz, D-78457 Konstanz, Germany — The nitrogen-vacancy (NV) center in diamond has emerged as a promising optically addressable qubit candidate, but optical methods are usually used only for spin initialization and readout through the defect's spin-dependent intersystem crossing (ISC) transition. Quantum dynamical control typically requires the application of microwave magnetic fields, limiting possible applications. Here, we demonstrate an all-optical method for unitary, arbitrary-axis spin control of single NV spins below 10 K based on stimulated Raman transitions<sup>2</sup>. Using our recently-demonstrated arbitrary-basis spin initialization and readout, we perform time-domain spin coherence measurements on single NV center spins solely with optical pulses. These techniques enable individual addressing of proximal NV center spins and could be used to probe other previously-inaccessible defect spin systems without ISC spin addressability.

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> B. B. Buckley Center for Spintronics and Quantum Computation, University of California, Santa Barbara, California 93106

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