Optical studies of ultrafast orbital dynamics of a single spin in diamond\textsuperscript{1} F.J. HEREMANS, D.J. CHRISTLE, C.G. YALE, D.D. AWSCHALOM, Institute for Molecular Engineering, University of Chicago, Chicago, IL 60637, L.C. BASSETT\textsuperscript{2}, B.B. BUCKLEY, Center for Spintronics and Quantum Computation, University of California, Santa Barbara, CA 93106, G. BURKARD, Department of Physics, University of Konstanz, D-78457 Konstanz, Germany — The nitrogen-vacancy (NV) center in diamond shows great potential as an optically addressable solid-state spin for use in quantum information and metrology. At low temperature ($T < 10$ K) the NV center’s orbital-doublet, spin-triplet excited state becomes stable and optically coherent with the ground state. Here we use ultrafast optical pump-probe techniques coupled with optical polarization selection rules to investigate coherent orbital dynamics of the NV center’s excited state\textsuperscript{3}. The experiments reveal dynamics which occur on nanosecond down to femtosecond timescales due to the interplay amongst these three orbital levels. These techniques enable all-optical control of the NV center’s spin state and could provide a probe to investigate orbital decoherence and phonon interactions in the NV center and other defect systems.

\textsuperscript{1}This work is supported by AFOSR, and DARPA.
\textsuperscript{2}Present address: Department of Electrical and Systems Engineering, University of Pennsylvania, Philadelphia, PA 19104