Abstract Submitted for the DAMOP14 Meeting of The American Physical Society

Phonon-Induced Coupling and Intersystem Crossing in Nitrogen-Vacancy Centers MICHAEL GOLDMAN, ALP SIPAHIGIL, STEVEN BEN-NETT, ALEXANDER KUBANEK, MIKHAIL LUKIN, Harvard University Nitrogen-vacancy (NV) centers in diamond have emerged as a versatile atom-like system, finding applications in both metrology and quantum information science. In both regimes, it is crucial to understand the interactions between the NV center's electronic state and vibrations in the diamond lattice. The NV center's broad appeal as a sensor – for nano-scale, bio-compatible thermometry and magnetometry, for example – hinges on our ability to initialize and read out the electronic state with a single non-resonant laser. Both of these mechanisms are the result of an inter-system crossing (ISC) into a metastable state, a phonon-assisted shelving process that has not been fully explained. Conversely, the NV center's appeal as a precisely controllable quantum register depends on our ability to resonantly excite either closed cycling transitions or closed lambda transitions. The fidelity of operations that depend on these transitions can be degraded by phonon-induced mixing of electronic orbital states within the excited state manifold, which can provide an unwanted non-radiative decay channel out of the desired subspace. We have directly measured population dynamics and decoherence in the excited state manifold. We have quantified the phonon-induced mixing rate and demonstrated that mixing can be completely suppressed at low temperatures. Further, we have measured the ISC rate for different excited states and developed a theoretical model that unifies the phonon-induced mixing and ISC mechanisms.

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Date submitted: 03 Feb 2014

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