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Optical Control of Nuclear Spin Ensembles in Diamond JONATHAN KING, JEFFREY REIMER, University of California, Berkeley — We present new results on the hyperpolarization of <sup>13</sup>C nuclear spins in diamond through optically-oriented nitrogen vacancy (NV-) defects. Optical illumination of high NVconcentration diamonds at cryogenic temperatures and 9.4 Tesla results in a negative nuclear spin temperature with measured bulk-average polarization over 5%, although local polarization may be higher. The negative spin-temperature is attributed to a population inversion within the dipolar energy levels of the NV- spin ensemble. In our quantitative model, nuclei near defects equilibrate with the NVdipolar energy reservoir and polarization is transported to the bulk material via spin diffusion. This model is tested by investigating a series of samples with varied defect density. We also investigate the nuclear hyperpolarization of NV- containing diamond nanocrystals. Such materials may be useful for surface transfer of polarization to target molecules for enhanced NMR sensitivity. Additionally, we investigate the dynamics and decoherence of the hyperpolarized nuclear spin ensemble and its interaction with electronic defect spins. Such phenomena are of fundamental interest to the use of diamond for quantum information applications.

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