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Near unity optical spin polarization of ²⁹Si nuclei in silicon carbide¹ W.F. KOEHL, A.L. FALK, P.V. KLIMOV, D.J. CHRISTLE, D.D. AWSCHALOM, Institute for Molecular Engineering, University of Chicago, K. SZÁSZ, V. IVÁDY, A. GALI, Institute for Solid State Physics and Optics, Wigner Research Centre for Physics, Hungarian Academy of Sciences — We demonstrate optical polarization of ²⁹Si nuclei that are coupled to the electronic spins bound at neutral divacancy and PL6 defects in 4*H*- and 6*H*-SiC. We polarize high concentrations (10¹⁶ cm⁻³) of nuclear spins and measure efficient nuclear polarization at temperatures ranging from 5 K up to room temperature. The peak polarization is near unity (99 ± 1%), corresponding to a 5 μ K effective nuclear bath temperature. To explain the large polarization, we locate the spin transitions of the defects' optically excited electronic states and show that nuclear orientation is strongest at the ground and excited state spin-sublevel anticrossings. These findings suggest that nuclear polarization in SiC is a powerful platform for quantum memories, nuclear gyroscopes, and probes for magnetic resonance imaging.

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