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Resonant optical spectroscopy and coherent control of Cr⁴⁺ spin ensembles in SiC and GaN¹

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Spins bound to point defects have emerged as an important resource in quantum information and spintronic technologies, especially as new materials systems have been developed that enable robust and precise quantum state control via optical, electronic, or mechanical degrees of freedom. In an effort to broaden the range of materials platforms available to such defect-based quantum technologies, we have recently begun exploring optically active transition metal ion spins doped into common wide-bandgap semiconductors. The spins of such ions are derived in part from unpaired d orbital electron states, suggesting in some cases that they may be portable across multiple materials systems. This in contrast to many vacancy-related defect spins such as the diamond nitrogen vacancy center or silicon carbide divacancy, which are formed primarily from the dangling bond states of the host. Here we demonstrate ensemble optical spin polarization and time-resolved optically detected magnetic resonance (ODMR) of the $S = 1$ electronic ground state of chromium (Cr⁴⁺) impurities in silicon carbide (SiC) and gallium nitride (GaN) [1]. We find that these impurities possess narrow optical linewidths (< 8.5 GHz at cryogenic temperatures) that allow us to optically resolve the magnetic sublevels of the spins even when probing a large ensemble of many ions simultaneously. This enables us to directly polarize and probe the Cr⁴⁺ spins using straightforward optical techniques, which we then combine with coherent microwave excitation in order to characterize the dynamical properties of the ensemble. Significantly, these near-infrared emitters also possess exceptionally weak phonon sidebands, ensuring that $> 73\%$ of the overall optical emission is contained within the defects' zero-phonon lines. These characteristics make the Cr⁴⁺ ion system a promising target for further study in the ongoing effort to integrate optically active quantum states within common optoelectronic materials. [1] W. F. Koehl, B. Diler, S. J. Whiteley, A. Bourassa, N. T. Son, E. Janzén, and D. D. Awschalom, arXiv:1608.08255.

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