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Charge State Equilibration of Nitrogen-Vacancy Center Ensembles in Diamond: The Role of Electron Tunneling

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Abstract

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The nitrogen-vacancy (NV) center in diamond is a well-established solid-state defect with promising applications ranging from quantum sensing of physical properties to room-temperature quantum computing. NV centers possess two charge states with well-established optical transitions: the neutral NV^0 state and the negatively charged NV^- state. The negatively charged state exhibits spin-optical properties that give the NV center its unique capabilities. In diamond crystals, single-substitutional nitrogen defects (N) are usually more abundant than NV centers and function as donors, providing NV centers with the additional electron required to attain their preferred charge state. During optical excitation of NV^-, photoionization can occur, promoting an electron to the conduction band and converting NV^- to NV^0. While optical protocols can restore NV^-, recovery is also observed in the absence of illumination, indicating a charge-transfer process from donors to NV centers.

Despite the central role of charge transfer in establishing and restoring the NV^- charge state, the microscopic mechanism enabling charge transfer between deep-level defects such as NV and N remains unresolved. In diamond, the high ionization energies of these defects rule out thermally activated carrier emission and capture as likely explanations. To address this long-standing issue, we use pump-probe spectroscopy to monitor the relaxation dynamics of NV^0 back to NV^- following photoionization. We find that following photoconversion of NV^- to NV^0, NV^- recovery strongly depends on the N donor concentration, with no discernible temperature dependence. These observations, supported by first-principles-based theoretical modeling, identify phonon-assisted electron tunneling between localized defect states as the dominant mechanism governing charge equilibration in NV center ensembles.