

Nitrogen and silicon defects in co-doped CVD diamond and their charge state behavior

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In most diamond for next-generation applications such as quantum information processing and magnetic sensing, significant efforts are made to avoid unwanted defects and uncontrollable charge transfer, with most material avoiding high doping regimes. Nevertheless, some low level of doping is typically advantageous. For instance, to stabilize ensembles of negatively-charged group-IV-vacancy (G4V) defects for ensemble quantum memory applications[1] or neutrally charged counterparts for single photon emission, it is critical to understand and control the defects' charge behavior to avoid coherence loss and fluorescence blinking. Despite boron-doped diamond's outstanding p-type conductivity, work to identify ideal n-type dopants in diamond is still ongoing. As the creation of n-type diamond by elemental substitution is proving difficult[2] (with only phosphorous showing any promise), co-doping approaches to create novel complexes such as BN₄, BN₂[3] and SiN₄ [4] have been theoretically explored and are predicted to be promising. Moreover, the understanding of charge transfer between novel, complex defects formed in co-doped CVD diamond is non-trivial.

In this work, several silicon and nitrogen co-doped CVD synthetic samples, covering multiple doping regimes, are comprehensively investigated. Annealing using temperatures in the range of 1600°C to 2400°C reveals the behaviors of silicon and nitrogen incorporation by tracking critical defects. When annealed at temperatures above 2000°C, several previously unreported optical features and EPR signals emerge. A set of EPR multiple lines with S=1/2 at approximately g = 2.004 are unambiguously assigned to a new defect, SiVN⁰[5], which anneals in above 2000°C. Several absorption and luminescence features are correlated with the production of several Si-N complexes. Further work to assign the unknown optical features to atomic structures are ongoing.

Co-doping of this material leads to N, Si, and H-related defects which possess shallow donor / acceptor levels, and hence charge transfer in these samples is significant. Static charge transfer and charge dynamics are observed in various defects such as SiV, N_s, NV, SiVN and NVH, etc. driven by bandgap UV irradiation and heat treatment (550°C) in ambient conditions. Moreover, the significance of photochromism caused by the charge transfer of SiV between its optically-active charge states and inactive one leads to unambiguous identification of SiV²⁻[5] which is EPR silent and has no optical features observed to date.

The complicated charge dynamics and defect annealing behavior enables the investigation of the spatial distribution of various defects, which is crucial to interpreting the charge behaviors and understanding defects' aggregation. High-resolution PL mapping reveals an anti-correlation spatial relationship between SiV and nitrogen-containing defects (NV and H3), which leads to an intriguing topic in exploring the growth mechanism of silicon and nitrogen co-doped CVD synthetic diamond.

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