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Accession Number

12263502

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Author Unabbreviated

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Title

Single substitutional nitrogen defects revealed as electron acceptor states in diamond using ultrafast spectroscopy

Source

Physical Review B (Condensed Matter and Materials Physics), vol.84, no.16, 15 Oct. 2011, 165202 (9 pp.). Publisher: American Physical Society, USA.

Abstract

We report on the carrier dynamics and recombination pathways of photogenerated electrons in type Ib synthetic diamond using ultrafast spectroscopic techniques. Samples with controlled amounts of nitrogen defects were grown using the high-pressure high-temperature (HPHT) method. Electrons were excited from single substitutional nitrogen defects into the conduction band via an ultrashort pulse from a frequency-doubled Ti-sapphire laser. Using time-resolved terahertz time-domain spectroscopy, we determined the mobility of the photoexcited electrons and monitored their recombination dynamics, at temperatures ranging from cryogenic temperatures to room temperature. The electron mobility was observed to be limited by scattering with neutral nitrogen impurity defects (N_s⁰). Electrons were observed to predominantly recombine into neutral nitrogen states rather than their original ionized nitrogen defects, thereby creating negatively charged nitrogen states (N_s⁻). The creation of N_s⁻ states is confirmed experimentally by tracking the localized vibrational modes (LVM) of nitrogen defects during the electron recombination process using visible pump-infrared probe transient spectroscopy. We observe a transient infrared absorption feature at 1349 cm⁻¹ that can be assigned to the LVM of N_s⁻. Density functional calculations are carried out to determine the LVMs of nitrogen in various charge states, and we find a ~10 cm⁻¹ upward shift of the mode on passing from N_s⁰ to N_s⁻, in agreement with experimental observations. (42 References).