Ab initio study of carbon defects in hexagonal boron nitride: in search of single-photon emitters

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Observation of single-photon emitters (SPEs) in hexagonal boron nitride (hBN) has rendered this material a promising platform for future quantum technologies. SPEs in hBN are observed in a wide spectral range, from near-infrared to ultraviolet, and many of them exhibit particularly promising optical and spin properties. However, the chemical identification of most of the observed emitters is still under debate. Experimental research provides a wealth of evidence that the presence of many of SPEs in hBN is correlated with the amount of carbon in the material, specifically referring to the presence of red, blue and UV emitters [1-3].

In this contribution we present summary of our *ab initio* studies concerning carbon-related defects in hBN and their potential relationship with observed SPEs. Particularly, we showcase results addressing three key aspects: (1) the thermodynamics of carbon point defects in hBN, (2) the origin of the red emission (ZPL at 1.6-2.1 eV), and (3) the origin of the blue emission (ZPL at 2.85 eV).

Regarding (1), we analyze thermodynamic properties of four types of carbon defects in hBN: (i) carbon complexes containing up to 10 atoms, (ii) carbon complexes with vacancies, (iii) carbon complexes with antisites, and (iv) carbon with oxygen atoms complexes. On the basis of calculated formation energies, equilibrium densities of defects are calculated for typical temperatures and growth conditions. The most prevalent of considered defects are carbon monomers (C_B, C_N), dimers, trimers, triangular (starlike) tetramers ($(C_B)_3C_N$, $(C_N)_3C_B$), and – in the presence of oxygen – C_NO_N. Carbon monomers prevail under extreme conditions of growth (N-poor, N-rich), but under intermediate condition dimer is the defect of the lowest formation energy. If oxygen is present and growth conditions are N-poor, the most abundant defect becomes C_NO_N. Complexes of carbon with vacancies and antisites appear at much lower densities as their formation energies corresponding to equilibrium Fermi level usually exceed 4 eV.

Regarding (2), the results of thermodynamic modeling are discussed in light of experimental results presented in [1]. For the defects deemed the most promising candidates, optical properties are analyzed. Monomers, trimers, and triangular tetramer defects are possible candidates for red SPEs. Triangular tetramers appear to be particularly interesting due to their triplet ground state. Pros and cons of these assignments, involving values of ZPL energies and symmetry, are discussed in the view of existing experimental data.

Regarding (3), we introduce a new model of the emitter based on carbon-related defects. Its calculated optical characteristics show excellent agreement with experimental observations. Specifically, the position of ZPL, radiative lifetime, electron-phonon coupling, and Stark shift align closely with experimental data [2-4]. Furthermore, the proposed model effectively explains experimental facts regarding the formation and annealing of blue emitters.

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