Coupling of excitons and defect states in boron-nitride nanostructures

C. Attaccalite, M. Bockstedte, A. Marini, A. Rubio and L. Wirtz

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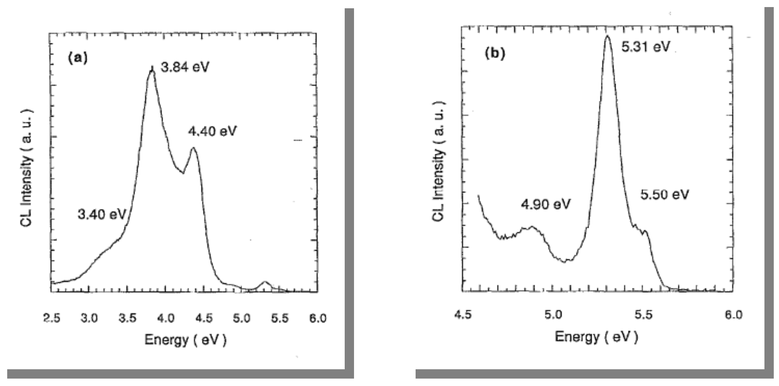
The signature of defects in the optical spectra of hexagonal boron nitride (BN) is investigated using many body perturbation theory. A single BN-sheet serves as a model for different layered BN-nanostructures and crystals.

The electronic properties of the hexagonal BN, in its different form, bulk h-BN, nanotubes, single sheet, are fundamental for developing deep ultraviolet light devices. Nowadays there are controversial experimental results on the luminescence from BN nanostructure and on its origin.



Deep-level and (b) near-band-gap W emission spectra measured at 4.2 K on a hexagonal BN film.

C. A. Taylor II, S. W. Brown, V. Subramaniam, S. Kidner, S. C. Rand, and R. Clarke
Appl. Phys. Lett., Vol. **65**, No. 10, 5 September 1994.

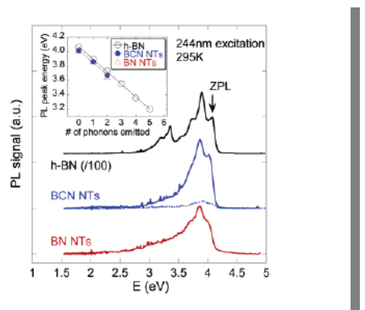


Figure 3. Continuous-wave photoluminescence spectra obtained at room temperature. For the $B_xC_yN_z$ NTs, the dashed curve shows the spectrum recorded when the excitation laser was focused on a dark grain in the sample. Inset shows the phonon replica energies plotted against the number of phonons emitted.

J. Wu, Wei-Qiang Han, W. Walukiewicz,, J. W. Ager III, W. Shan, E. E. Haller, and A. Zettl NANO LETTERS 2004 Vol. **4** 650

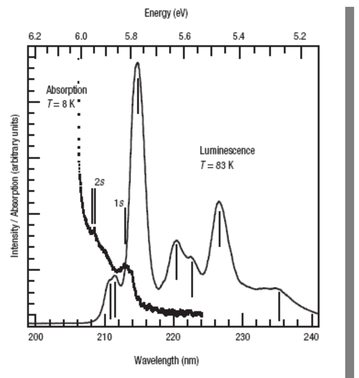


Figure 2 Fundamental absorption spectrum at 8 K and luminescence spectrum at 83 K. The weak fringes due to interference between the front and back surfaces are observed from 213 to 224 nm in the absorption spectrum. The bars indicate the peak positions for each spectrum.

KENJI WATANABE, TAKASHI TANIGUCHI AND HISAO KANDA
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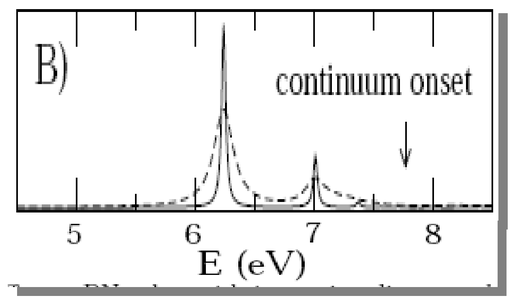
From a theoretical point of view it has been shown that the optical absorption of BN is dominated by strong bounded excitons and is slightly effected by the crystal orientation of the BN planes

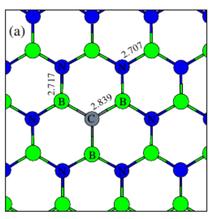
Optical absorption of a single sheet of hexagonal BN calculated using GW + Bethe Salpeter



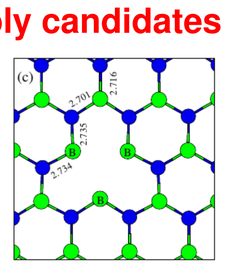
Ludger Wirtz, Andrea Marini, and Angel Rubio
Phys. Rev. Lett. 96, 126105 (2006)

Are the Luminescence spectra due to electron-hole recombination on defects?

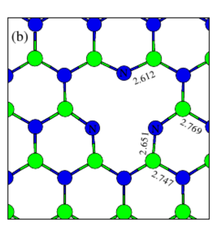
The most probably candidates are:



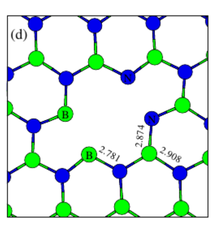
1) Carbon substitution of a Nitrogen
acceptor impurity



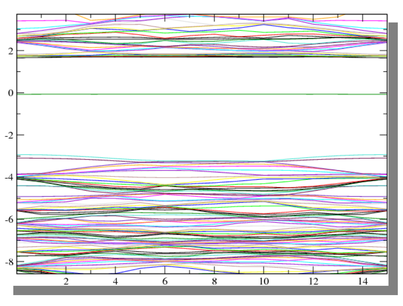
2) Nitrogen vacancy
donor impurity



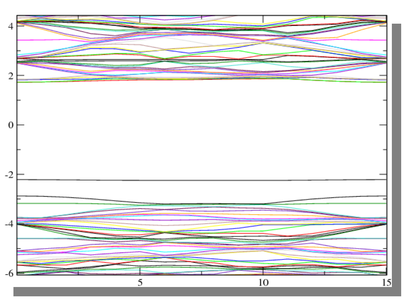
3) Boron vacancy
acceptor impurity



4) Boron-nitrogren vacancy

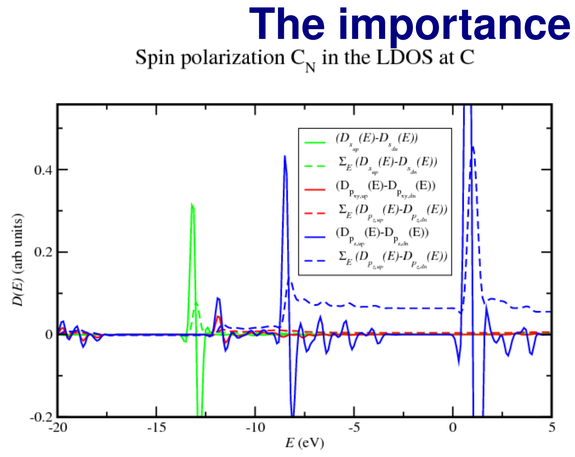


Defects introduce deep level in the band gap



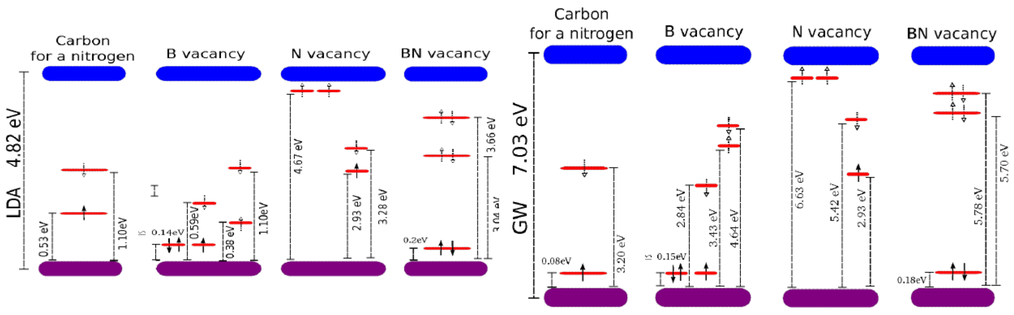
Contact: claudio.attaccalite@grenoble.cnrs.fr

The importance of spin polarization



A large class of impurities when introduced in h-BN aquires are in a spin-polarized state with a local magnetic moment. This is show for example for the carbon impurity by plotting the project local density of states. The effect is not small and implies that the Green's functions of the two spin-channels differ significantly.

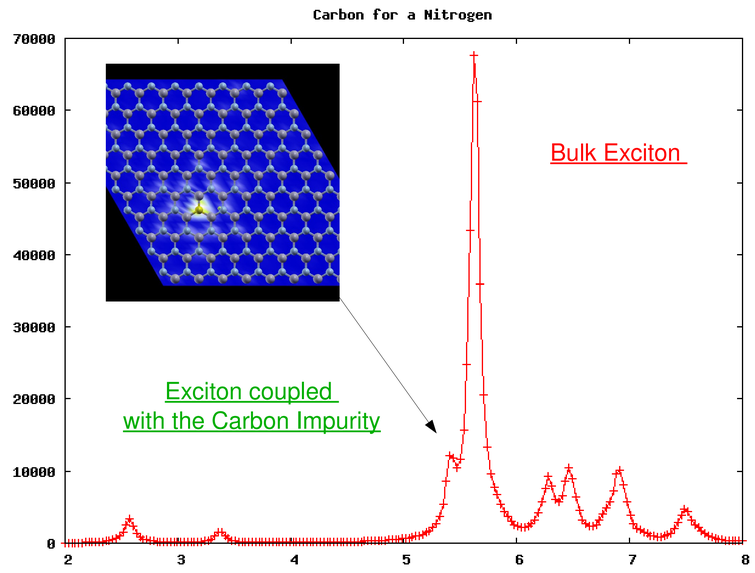
Quasi-particle levels within G_0W_0

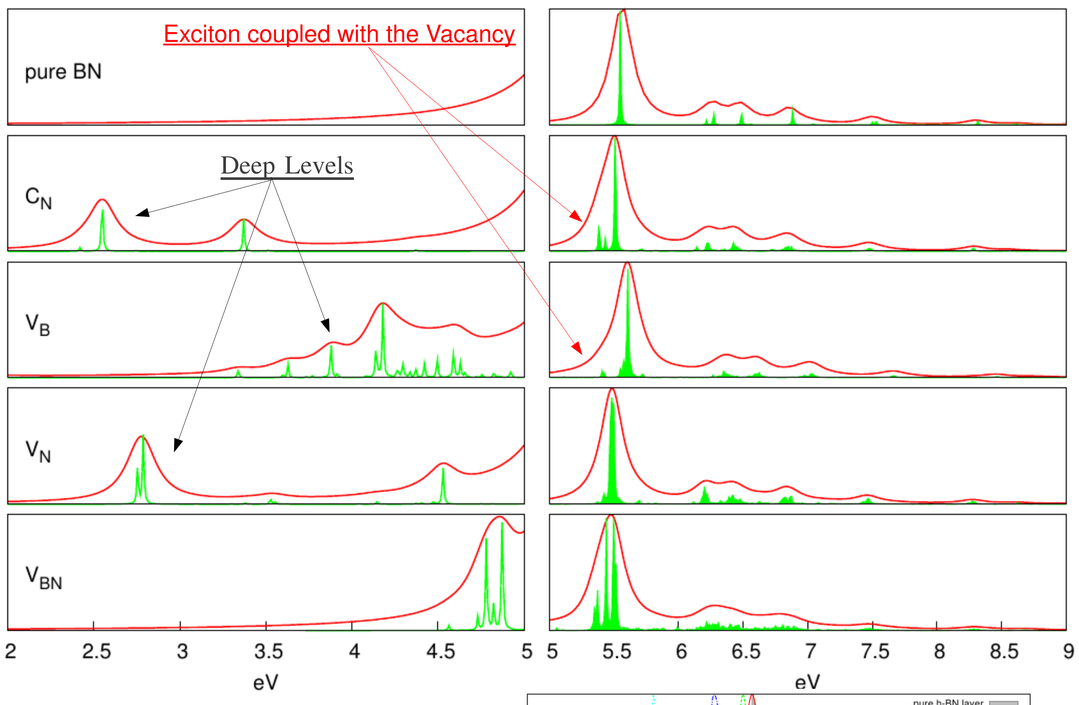


Quasiparticle energies of C_N , V_B , V_N , and V_{BN} as obtained from LDA and G_0W_0 calculations: schematic representation of energy position with in the band gap. The filled arrows indicate levels occupied with electrons with spin up or down. Dotted arrows indicate unoccupied states (holes) correspondingly.

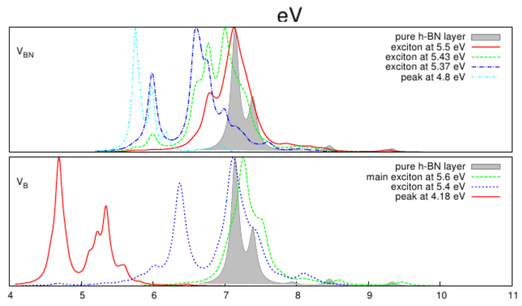
Optical absorption

We calculated the optical spectra by Bethe-Sapeter equation in presence of defects, here below the result for C_N:





Electron-Hole pairs that contributes to Excitonic State



Conclusions

We found that transitions between defect levels give rise to the deep peaks in the absorption while the coupling between defect and bulk levels generates the peaks just below the main exciton. In the case of di-vacancy the reduced symmetry splits the exciton in three sub-peaks.

These results can explain the luminescience spectra around 4eV and the splitting of the main exciton in the emission spectra.