Elementary excitations of quantum emitters in highly defective hexagonal Boron Nitride

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The recent discovery of bright and tunable quantum emitters (QEs) in hexagonal Boron Nitride (hBN) stable up to room temperature enables the realization of novel and scalable quantum photonic platforms [1,2]. While it is well-accepted that SPEs in hBN stem from defects [3], key details on their origin, electronic levels, and orbital involvements are still unknown. Here we interface, for the first time, measurements with resonant inelastic X-ray scattering (RIXS) and photoluminescence (PL) on pristine and plasma-treated hBN with high density of defects. RIXS measurements performed at the π* antibonding orbitals of Nitrogen uncover a fundamental excitation E_0 at 285 meV that generates harmonics with energy $E_n=nE_0$ (n=1,2, 3,...) ranging from the mid-IR through the UV (Fig.1a,b). These fundamental harmonics are observed only in highly defective samples and not in pristine samples. PL spectroscopy at low temperature indicates that highly defective samples host several QEs (Fig.1c). We analyse the QE emission pattern with a model that accounts for donor-acceptor-pair (DAP) recombination process. In the spectral range covered by our PL measurements, the DAP fit shows the presence of four transitions with energy overlapping the one of the harmonics with n=5,6,7,8 (Fig.1d). The correlation between the harmonics and the DAP transitions indicates that most of the QE observe in hBN have a common origin that can be ascribed to the N π* orbitals. Our interpretation generalizes QEs in the IR, visible, and UV range through a single energy scale explaining the stability and robustness of SPEs in hBN. Moreover, due to the orbital sensitivity of RIXS, we can underscore the association of QEs in hBN with the π* antibonding orbitals.

References

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Figures

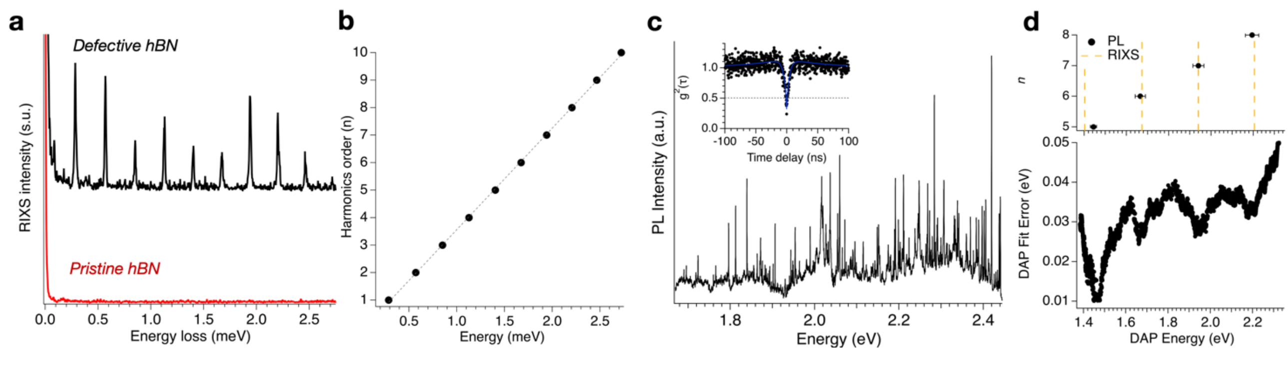

Figure 1: a – RIXS spectra taken at the N K pre-edge (π*) of defective (black curve) and pristine (red curve) hBN. **b** – Energy of the harmonics extracted from **a**. **c** – PL spectrum of defective hBN at T=5 K. Several sharp peaks indicate the presence of many QEs. The inset show an example of second-order correlation function ($g^2(\tau)$) measured on an individual line. Antibunching below 0.5 confirms the single-photon emission. **d** – Results of the DAP fit of the energy of the QEs measured with PL experiments. Four DAP transitions result from the fit as their energy minimize the fit error. The top panel shows the comparison between the energy of the DAP transitions and RIXS harmonics for n=5,6,7,8.