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Low-temperature spectroscopy of single-photon emitters in irradiation-engineered hexagonal boron nitride

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Abstract: To identify the microscopic origin of single-photon emitters in hexagonal boron nitride, we perform low-temperature spectroscopy. We observe strong photoluminescence at two different excitation lasers that hint at a phonon-assisted excitation scheme.

Research on the two-dimensional material hexagonal boron nitride (hBN) increased strongly in 2016 when single-photon emitters (SPEs) at room temperature were discovered [1]. However, the microscopic origin of these SPEs remains elusive and the generation mechanism is unclear for many processes. Here we present a novel generation process to generate luminescent centres in hBN [2]. We exfoliate high-quality hBN flakes and irradiate them with oxygen atoms, as sketched in Fig. 1a. After subsequent annealing in nitrogen, we find several single-photon emitters, confirmed by second-order correlation measurements [2].

We systematically study the density of luminescent centres at different irradiation energies and fluences (defined as the number of oxygen atoms per area). Increasing the irradiation fluence by ten results in a five-fold enhancement of the density. In combination with molecular dynamics simulations we clarify the generation mechanism, for the first time to the best of our knowledge. Furthermore, we infer that most likely two types of defects are generated [2]. One of these defects is a nitrogen vacancy next to a carbon atom on a boron site, labelled as $V_{\text{N}}\text{C}_{\text{B}}$. Ab initio calculations of this defect show excellent agreement with experimental photoluminescence, as shown in Fig. 1b. The generation of quantum emitters by irradiation engineering is a step towards the controlled generation of quantum emitters in hBN. Furthermore, the presented irradiation engineering is wafer-scalable and could be adapted to other irradiating atoms or ions as well as other gapped two-dimensional materials.

The experimental photoluminescence line shape in Fig. 1b shows a narrow zero-phonon line (ZPL) associated with pronounced red-shifted phonon side bands (PSBs). We ascribe such a line shape to luminescent centres of Group I. To investigate the PSBs of Group I in more detail, we carry out low-temperature measurements as shown in Fig. 1c. The top panel shows a room and low temperature spectrum under excitation with a green laser at a wavelength of 523 nm. It reveals individual peaks in the PSBs that are unresolved at room temperature. In the field of single-photon emitters in hBN, blue lasers are only rarely used for excitation. In the bottom panel of Fig. 1c we show room and low temperature photoluminescence of a Group I centre under blue excitation at 488 nm. As for green excitation, we observe highly-resolved PSBs at low temperature.

In Fig. 1c, the ZPL energy under green excitation is red-shifted by 140 meV from the laser energy. Furthermore, the energy difference between the blue and the green laser is 170 meV which is close to optical phonons in hBN with energies from 160 to 200 meV [3]. From these findings we infer that the excitation of Group I emitters is mediated by optical phonons as proposed in previous reports [4,5].

Fig. 1d shows the corresponding Jablonski diagram where the PL emission is strong when the laser detuning is equal to multiples of the phonon energy, e.g. $\hbar\Omega$ and $2\hbar\Omega$ where $\hbar\Omega$ is the phonon energy. To verify this phonon-assisted excitation scheme, we perform further measurements at different laser energies on the same Group I centre. We will also discuss how such measurements can be used to shed light on the microscopic origin of SPEs in hBN.

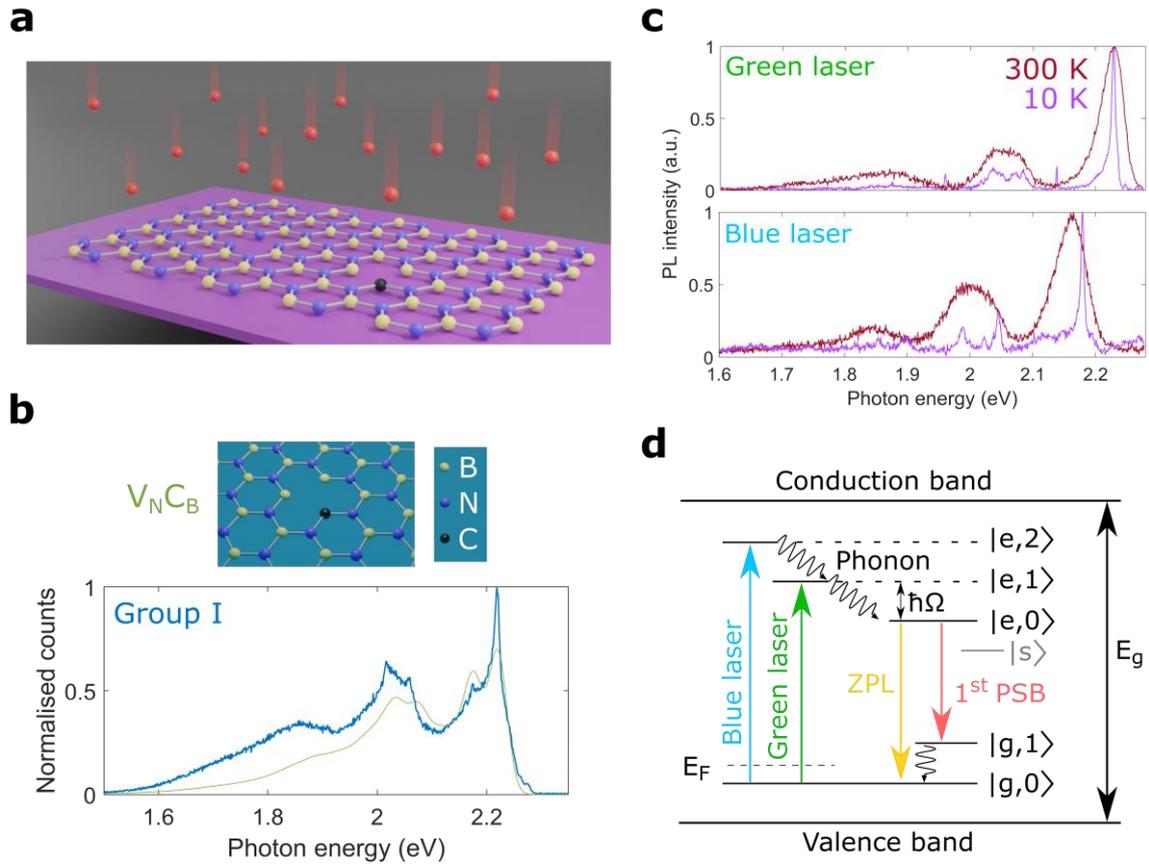


Fig 1. Spectroscopy of luminescent centres (LCs) in hBN. (a) Oxygen irradiation of hBN. Oxygen atoms are shown in red while boron and nitrogen atoms are shown in yellow and blue, respectively. One $V_N C_B$ defect is shown in the hBN lattice where the carbon atom is shown in black. (b) Comparison of experimental and theoretical photoluminescence (PL) line shapes. The top panel shows a sketch of the $V_N C_B$ defect in the hBN lattice. The bottom panel shows the experimental line shape in blue and the theoretical curve in green. (c) Room and low-temperature PL is shown under green and blue excitation in the top and bottom panel, respectively. The zero-phonon line (ZPL) energies are at 2.23 eV and 2.18 eV under green and blue excitation, respectively. The narrow peak around 2.10 eV under green excitation is assigned to another emitter in the same excitation spot. (d) Jablonski diagram of LCs in hBN. The PL emission of the LC consists of a ZPL and phonon sidebands (PSBs). For clarity, only the first-order PSB (1st PSB) is shown. The PL is strong for laser detunings equal to $\hbar\Omega$ and $2\hbar\Omega$ where $\hbar\Omega$ is the phonon energy. The Fermi level is labelled as E_F and the band gap is $E_g \approx 6$ eV [6].

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