Laser-related Spectroscopic Parameters of NV Colour Centres in Diamond

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One of the most common impurities in synthetic diamond is single substitutional nitrogen, which is incorporated in the diamond lattice substituting a carbon atom [1]. If the nitrogen is adjacent to a vacancy in the diamond lattice, it forms the nitrogen-vacancy (NV) colour centre (CC) [1]. The negatively charged state of this CC, NV-, is particularly well studied since its quantum properties are suitable for applications such as quantum information processing, single-photon sources and optical magnetometry [2]. NV CCs in the neutral state (NV0) are less widely studied. This CC exhibits broadband luminescence at slightly shorter wavelengths than NV-, and hence is also potentially of interest for tuneable and ultrafast visible laser applications. In this report, we present a detailed study of the laser-related spectroscopic properties of a diamond containing NV0 and NV- CCs.

The synthetic diamond studied was grown by chemical vapour deposition by Element 6. After electron beam irradiation (electron density of $1.6\text{x}10^{17}$ e/cm^2 and energy of 4.5 MeV) and 5 hours annealing (temperature of 800 °C) concentrations of NV0 / NV- CCs of 0.19 ppm / 0.5 ppm were measured. The absorption spectra of the sample before and after treatment are shown in Fig. 1 (a). Narrow peaks in the spectrum after treatment at 575nm and 637 nm are associated with the zero phonon lines of NV0 and NV- CCs [2]. The luminescence spectra of the diamond (Fig. 1(a), left inset) are affected by the balance of CCs excited by a particular pump wavelength. For 532 nm pumping, both NV0 and NV- CC are excited, and both H3 (two substitutional nitrogen atoms adjacent to a vacancy) and NV0 CC are excited at 450 nm. Emission from H3 CC can be seen as a characteristic shoulder in the spectrum at ~510 nm. The typical luminescence decay kinetics (Fig. 1 (a), right inset) for the spectral range > 640 nm is mono-exponential (decay time of 20-22 ns) for 448 nm pumping, and double-exponential (fast decay time of 8-9 ns and slow decay of 19-21 ns) for 562 nm pumping. We attribute the difference in the luminescence decay kinetics at pump wavelengths of 448 and 562nm to different CCs being excited: mainly NV0 at 448 nm and both NV- and NV0 at 562 nm. The radiative lifetime of NV0 CC is believed to be 19±2 ns. The reported radiative lifetime for NV- is 13±0.5 ns. Based on these values, the quantum yield QY of NV- and NV0 CC in the sample can be estimated to be ~65±4% and ~100%, respectively. The stimulated emission cross sections of the NV CCs were calculated using the Füchtbauer-Ladenburg equation (Fig. 1 (b)) taking into account the radiative lifetime of NV0 and NV- CC of 20 and 13 ns, respectively. To do that the luminescence spectra of the sample pumped at 450 and 532 nm were corrected for the contributions from H3 and NV0 CC. The peak emission cross-section of NV- CC was calculated to be $\sigma_{em}=3.5\times10^{-17}$ cm^2 at 715 nm, and that of NV0 CC is $\sigma_{em}=1.6\times10^{-17}$ cm^2 at 650 nm. The gain spectra (Fig. 1 (b), inset) was calculated from the emission and absorption spectra of the sample for different inversion factors β. The calculated gain for NV- CC is positive in the spectral range between 670 and 820 nm for β = 0.3, which makes it a promising candidate for laser action. The gain in NV0 is somewhat lower. The gain coefficient is positive only at β>0.3, with a maximum value of <0.5 cm^{-1}.

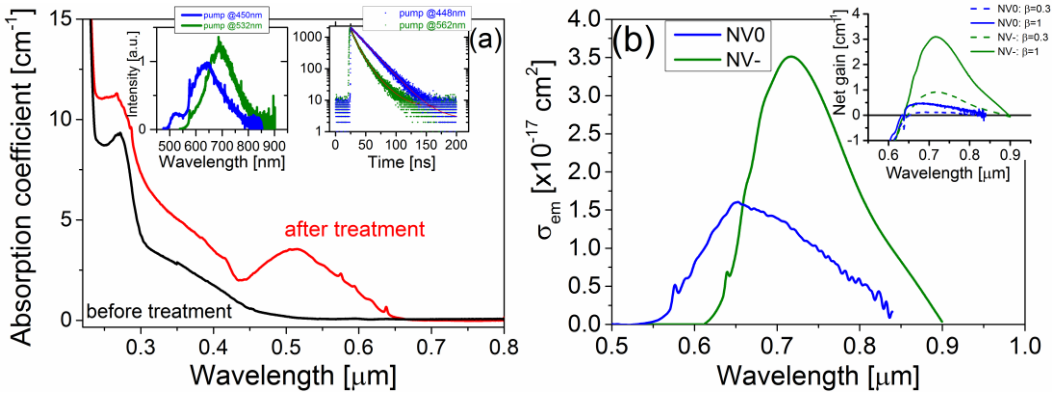

Fig. 1 (a) Absorption spectra of the sample before and after the treatment. Insets: luminescence spectra (left) and luminescence decay kinetics (right) pumped at ~0.45 and ~0.53 μm. (b) Emission cross-section of NV0 and NV- CCs. Inset: Calculated gain spectra of NV- and NV0 CC at the pump wavelength of 0.53 μm (green) and 0.45 μm (blue) for different inversion factors β.

References

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