Joint phasor analysis and iterative fitting of lifetime data in synthetic diamond

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Abstract

In this poster, a previously developed multidimensional luminescence spectrometer and microscope is applied to a synthetic diamond, consisting of a HPHT substrate and CVD grown layer. Hyperspectral imaging under 500 nm excitation reveals NV and SiV^- defects with varying concentrations over the sample. Lifetime imaging at two emission bands, with decay analysis using joint phasor and iterative fitting is also presented . Spatially varying spectroscopic analysis indicates quenched emission of NV^- in the HPHT substrate. This quenching is found to be both spatially varying and excitation wavelength dependant.

Diamond contains many luminescent optical centres (defects) which are optically active across a broad spectral range. In addition to this large number of optical centres, the lifetime of the excited state can vary significantly between different defects. This can also extend to lifetime variations of the same defect in a given sample. This can occur due to quenching of the defect, allowing the excited electron to decay through an additional route. The effect is a reduced average lifetime, and a reduction in quantum efficiency caused by the (often non-radiative) pathway introduced by the quenching species. Whilst typical defects in diamond that exhibit nanosecond fluorescence emission can often be accurately modelled by a single exponential decay per species, a quenched emitter can only be approximately fitted by a multiple exponential model.

This can be challenging since fitting to a complex exponential decay can be highly sensitive to the parameters entered into the model, including the initial guess and number of components. Spectroscopy of diamond at room temperature results in broad phonon side bands for a variety of common defects such as NV, which can obscure weaker emitters and result in a spectral overlap in luminescence information. Instead of emission wavelength, the lifetime components can be used as a contrast mechanism to reveal the weaker components. Literature can provide the initial values for an unquenched emitter using spectroscopic methods for the defect's identification, but 'hidden' components cannot have their lifetimes extracted this way. This due to the lack of identifiable structure in the spectrum for identification, such as ZPLs.

Phasor analysis is an alternative method for lifetime analysis and uses a direct Fourier transformation of the decay data to reveal different components. The resulting real and imaginary values are plotted on a phasor plot, which provides a visual graphical method to extract lifetime values. These are not prone to the same initial guess errors as iterative nonlinear fitting. The values from the phasor plot can then be used in fitting procedures to provide the initial guesses, thereby improving the fitting methodology. Complex decays, such as quenched decays can be analysed using phasor techniques, providing phase and modulation lifetimes that can be considered as representative "average" lifetime values for the luminescence. Phasor analysis can be used in conjunction with complex spectroscopic techniques, such as excitation-emission-lifetime matrices to reveal these quenched emitters.