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Creation and charge state dynamics of nitrogen-vacancy colour centres in diamond for quantum applications

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ABSTRACT

In the past decades the negative nitrogen-vacancy (NV^-) centre in diamond has demonstrated its versatility both as a sensor for temperature, electrical and magnetic fields, and as a promising solid-state system for quantum information processing. Potential applications of NV centres strongly depend on the quality of the initial diamond host material and on the NV's creation process itself, as it directly influences the NV's local environment and thus its properties. Limiting decoherence effects can be attributed to factors including the spin bath of ^{13}C atoms, electron donors (e.g. nitrogen) and crystal damage.

One part of this PhD thesis describes the creation of NV centres tailored for different applications: Neutron versus electron irradiation for dense ensemble creation in nitrogen-rich diamond crystals, and low energy nitrogen implantation (8 keV) for precisely positioned NV centres in ultrapure diamond, within a thin layer just 12 nm below the surface. In the nitrogen implantation series we created shallow NV ensembles of different densities and studied the influence of implantation dose (10^{10}-10^{14} /cm² nitrogen ions) on the NV's properties. However, long-lived spin coherence of NV^- would require at least a stable charge state, but it is photochemically not stable and thus loses the essential spin characteristics as soon as it converts to the neutral charge state, NV^0. The charge state dynamics therefore play a key role in the fundamental understanding and application of the color centre. In one of the key investigations of the thesis, we explored the time dependence of de-ionization ($NV^- \rightarrow NV^0$) and recombination ($NV^0 \rightarrow NV^-$) for shallow NV centres of different densities via a series of pulsed multi-color laser excitation sequences. In addition to laser-induced processes, we studied the spontaneous charge conversion and discovered that the characteristics of the decay ($NV^- \rightarrow NV^0$) depend strongly on the density of the color centres. This is of importance for numerous applications of NV centres, e.g. for sensing charges and fields near the surface of the diamond, as the decay process limits the coherence time – and thus the sensitivity – of NV-based sensors.

Aiming towards Purcell-enhanced NV centres inside an array of microcavities, we also explored the potential of masked implantation via electron-beam lithography and subsequent nitrogen implantation to create arrays of implanted NV defects. Fine-tuning the initial mask resist parameters and the implantation fluence has enabled us to scale NV creation down to single NV centres. We furthermore fabricated nitrogen implanted diamond samples with gold electrodes directly on the diamond surface. These devices were used to perform photocurrent detection of magnetic resonance (PDMR), a new method to read out the NV^- spin states electrically on a diamond chip. The nanometer-scale accuracy of masked implantation, combined with the fabrication of electrical and microwave structures on the diamond surface, will allow the creation of large-scale arrays of NV-based sensors and quantum devices.