Exploring nonlinear optical effects in NV-doped diamond

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The optical properties of negatively charged nitrogen-vacancy (NV^-) centers in diamond have garnered significant attention due to their potential applications in biology, medicine, quantum sensing and quantum information processing. In most cases, visible to near-infrared luminescence is recorded, upon the green laser excitation, and carries information on the NV spin which, in turn, may be manipulated using microwave fields. Although there is some research on nonlinear optical effects in bulk diamond and nanodiamonds, the impact of NV centers on such phenomena is far from being fully understood.

Here, we investigate the influence of NV centers in high-quality crystalline diamonds (DNV-B1, B14, and electronic grade; Element Six / Thorlabs) on nonlinear optical effects using 230-fs-long laser pulses with a wavelength of 1030 nm and a repetition rate of 1 kHz. In particular, the optical Kerr effect (OKE) and multi-photon absorption were explored using the Z-scan technique. Our findings reveal that diamonds with NV centers generally exhibit a lower non-linear refractive index compared to pure diamond. However, unlike pure diamond, the NV^- diamonds exhibit very strong nonlinear absorption effects, attributed to its bandgap energy level structure. As the energy of the laser pulse increases, we observe two-photon absorption (TPA) initially, followed by saturated absorption (SA), and finally reversed saturated absorption (RSA). These effects are also strongly enhanced with increasing NV concentration.

To identify the underlying processes, fluorescence (FL) spectra excited by laser pulses of various energies were examined. These spectra consist of a zero-phonon line (ZPL) of NV^0 observed at 575 nm and a broad band centered around 640 nm, which are associated with electron transitions between excited and ground states of NV^0. The lack of NV^- ZPL at 637 nm, a prominent feature under green light illumination, indicates the conversion of NV^- to NV^0 caused by multiphoton ionization. The spectrally integrated FL follows a quadratic dependence on laser pulse energy indicating the involvement of two-photon processes in the excitation of the FL spectra.

In summary, our studies unveil intriguing optical nonlinearity in diamonds with NV concentrations of a few ppm, which holds significant interest for researchers in the field of quantum sensing.

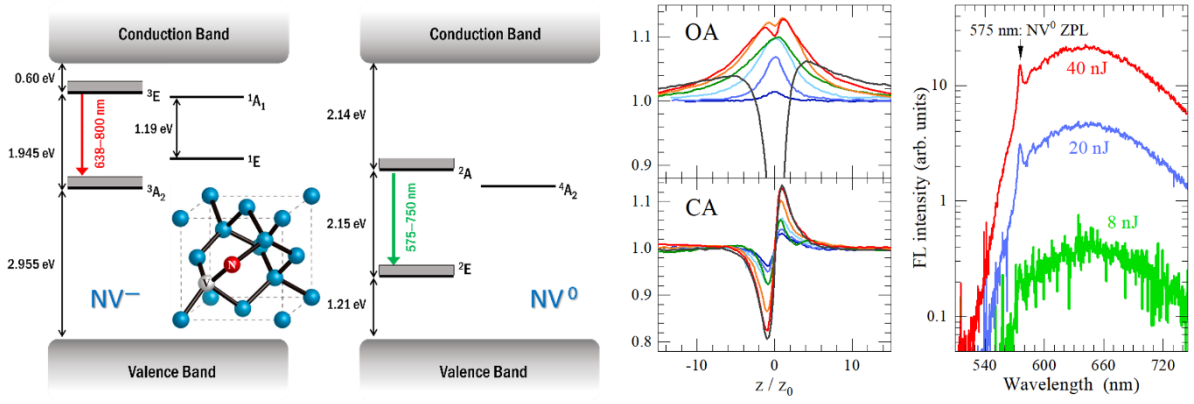

Figure: (from left to right) Bandgap energy level structure of NV-doped diamond. The normalized open-aperture (OA) and closed-aperture (CA) Z-scan signals recorded for laser pulse energies varying from 500 nJ to 9 µJ. Fluorescence spectra excited with laser pulses of different energies.