**Optically detected magnetic resonance study of thermal effects due to absorbing environment around nitrogen - vacancy - nanodiamond powders**

**Mona Jani,1,2,3,a) Zuzanna Orzechowska,1 Mariusz Mrózek,1 Marzena Mitura-Nowak,4 Wojciech Gawlik1 and Adam M. Wojciechowski1,a)**

**AFFILIATIONS**

**1**Marian Smoluchowski Institute of Physics, Jagiellonian University, 30-348 Kraków, Poland

**2**School of Engineering and Applied Science, Ahmedabad University, 380009 Gujarat, India

**3**Laser Centre, University of Latvia, Jelgavas street 3, LV-1004, Riga, Latvia

**4**Institute of Nuclear Physics, Polish Academy of Sciences, Radzikowskiego 152, 31-342 Kraków, Poland

**a)**Author to whom correspondence should be addressed: mona.jani@lu.lv and a.wojciechowski@uj.edu.pl

**SUPLEMENTARY MATERIAL**

**Photoluminescence spectroscopy:**

PL of NV centers in NDs was observed with a homemade confocal microscope with green 532 nm laser excitation at constant output power (below 0.5 mW to avoid laser heating). The PL properties of the NV centers in ND are affected mainly by the size of the particles and the surface modification1–3. Specifically, for an irradiated diamond, the graphite-related defects on the diamond surface may form electron traps where donor ionization and conversion of the charge state of the NV defects charge state may take place4. This conversion depends on the presence of impurities and other defects5–7, in particular Fe ions.

In the recorded PL spectra (Fig. S1), the most important features are the 576 nm and 637 nm peaks which are zero-phonon lines (ZPLs) of the NV0 and NV− centers, respectively. They are accompanied by broad phonon sidebands that extend to about 800 nm and peak at around 700 nm. The shape of the luminescence spectra is moderately changed as a consequence of the implantation of Fe inside NDs. The presence of 575 and 637 nm peaks confirms the presence of a considerable number of NV0 and NV− color centers in the investigated NDs. Slight changes in the NV−/NV0 ratio for the Fe-NDs are observed as shown in the inset in comparison to a pristine NDs. Since the overall brightness of NDs is the main criterion for comparison with other biomarkers, we conclude that the Fe-implantation does not impact the luminescence of the NDs in a meaningful way.

**Spin-relaxometry:**

Relaxation times were measured in the confocal microscope as previously described 8. The microwave pulses of 2.87 GHz frequency were produced by a controllable-programmable pulse generator (SRS SG396, Stanford Research Systems, US), fed to a high-power amplifier (ZHL-16W-43+ Mini-Circuits, US), and delivered to a loop-gap type antenna structure on a printed circuit board. The investigated ND and Fe-ND samples were placed directly on the antenna board.

We focused on spin relaxometry, i.e. measurements of the spin-lattice and spin-spin relaxation times, *T*1 and *T*2, respectively. Such measurements with NV− centers serving as a spin probe enable studies of the incorporation of Fe ions into NV diamond material. The *T*1 and *T*2 measurements were based on the “relaxation in dark”9,10 and “Hahn-echo”11 methods, respectively. Briefly, the applied relaxation in a dark method is a common-mode rejection procedure, which cancels most of the background fluorescence except that from the chosen NV− sub-ensemble (inset Fig. S2a). This allows one to measure only the time-dependent fluorescence from the NVs in a chosen crystallographic orientation and reject contributions from the other ones. The measurements start with the initialization of the spin state with a laser and application of a resonant microwave π pulse. Then the spins relax in the dark over time τ after which they are again interrogated by light. Next, the identical signal yet without the π pulse is recorded and subtracted. Taking a series of such measurements for various times τ enables recording of the time dependence of the NV fluorescence amplitude, fitting it with an exponential curve, and retrieving the value of *T*1 relaxation time. The Hahn-echo method relies on the application of three microwave pulses (inset Fig. S2b). After the spins are prepared to superposition state by the first microwave pulse of π/2 duration, a second π pulse is used to invert the spin phases before using the third π/2 pulse. Such a procedure reduces much of the spin dephasing and enables accurate measurements of the spin coherence times.

Fig. S2 shows the relaxation rates 1/*T*1 and 1/*T*2 measured in the NV− centers in ND and NDs with various implantation fluences. The optical and microwave pulse sequences used are depicted in the insets. The length of the microwave π pulses in these measurements was chosen to be 220 ns, and the laser power was 1.5 mW. Application of the common-mode rejection procedure described above yields the relaxation rates 1/*T*1 of the pristine ND and NDs implanted with various Fe-doses and Fig. S2a depicts the measured values for *B* = 0 and *B* = 4.1 mT magnetic field intensities. The magnetic field intensity of 4.1 mT is large enough to resolve the ODMR spectral features of NVs with various orientations that occur at lower magnetic fields. The graph shows that the *T*1 relaxation time for ND is 2.99 ± 0.42 ms which is in agreement with previous reports12. With the highest applied implantation dose, the *T*1 time is reduced to 0.62 ± 0.03 ms, which is an order of magnitude lower than for NV centers in ND. The graph also shows that the 1/*T*1 relaxation rates are dependent on the applied magnetic field, and the measured *T*1 values increase slightly with the field.

The measurements of *T*2 relaxation times of NDs implanted with magnetic ions (Fig. S2b) are not as often reported in the literature as the *T*1 ones. However, the spin-spin relaxation time *T*2 associated with the decay of spin coherence is intrinsic to the NV center and may provide important information on spin couplings with the environment. The *T*2 relaxation times depend on the particle size and the physical environment which can include modifications of the diamond sample induced by strain and/or temperature, and concentration of impurities (the substitutional nitrogen being the most important) plus surface spins as possible sources of decoherence13,14. Measurement of *T*2 relaxation timesin NDs at room temperature and for B = 0, yields the *T*2 relaxation time of ND as 1035.9 ± 84.7 ns. For the highest Fe implantation dose, *T*2 reduces to 256.2 ± 71.4 ns. We have also observed enhancement of the relaxation times by applying magnetic fields.

We attribute the reported here reduction in spin relaxation time to the additional relaxation channel opened by the Fe-ion spins in the ND matrix. Similarly, as in the case of *T*1, and *T*2, the additional relaxation mechanism depends also on the magnetic field and can be partially mitigated by the application of a magnetic field.

**ODMR (after annealing):**

Fig. S3 is the ODMR spectra of ND14 and ND15 before annealing collected (a, b) at a laser power of 65 mW at various MW powers and (c, d) with a MW power of -20 dBm at various laser powers. The figure shows that after annealing the samples, they represent the ODMR spectra similar to those of the NDs.

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**Fig. S1.** PL spectra of the Fe implanted 140 nm-sized ND with various implantation doses at 532 nm laser excitation. Inset shows amplitudes of the NV0, NV­− peaks and their ratios NV−/NV0 obtained from the average of ten PL spectra.





**Fig. S2.** (a) Longitudinal relaxation rates 1/*T*1 and (b) transverse relaxation rates 1/*T*2 at *B* = 0 (black squares) and 4.1 mT (red squares) for various doses of Fe-implantation. The insets illustrate the timing of the optical (green) and microwave (blue) pulse sequences for (a) *T*1 and (b) *T*2 measurements. Each measurement was repeated ten times at various locations on the sample, and the values were averaged to obtain the relaxation rates with error bars.







**Fig. S3.** ODMR spectra of ND14 and ND15 after annealing collected (a, b) at a laser power of 65 mW at various MW powers and (c, d) with a MW power of -20 dBm at various laser powers. The insets in (c, d) shows the zoomed-in image of the ODMR central resonance frequency.