Peculiarities of optical and ODMR Spectra of Nitrogen–Vacancy Color Centers in Diamond Crystals

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Peculiarities of optical and ODMR Spectra of Nitrogen-Vacancy Color Centers in Diamond Crystals

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Abstract. The optical and ODMR spectra of nitrogen vacancy (NV) centers in diamond were investigated as applied to the development of micro-scale diamond magnetometer sensor. It is shown that the NV^0 center (unlike the NV^- center) has an absorption line in the UV range. The inversion of the zero phonon line of the NV^- center was observed and explained. ODMR signals in high magnetic fields (as compared with inner fields in the crystal) as well as in weak fields also were investigated. We propose the solution to the problem of the magnetometer sensitivity reduction at low magnetic field, that is of interest for quantum magnetometry applications.

1. Introduction

NV centers in diamond crystals are of enormous interest for modern quantum optics, informatics, cryptography, and magnetometry. First of all, it concerns the negatively charged NV^- centers by virtue of the possibility of excitation and observation of ODMR (optically detected magnetic resonance) in them [1]. In this work we took the first step to the application of these centers to the task of precision measurement of sub-millimeter-scale magnetic fields by investigating their photoluminescence and ODMR spectra.

2. Experimental investigation of the luminescence spectra of NV centers in diamond

Photoluminescence spectra were recorded for two specimens of diamond, which were provided by the Lebedev Physical Institute of the Russian Academy of Sciences. Specimens were prepared at that institute as follows: synthetic diamonds (from Element Six; type SDB 1085 60/70; crystal volume, ~0.01 mm3) were irradiated by electron beams with intensities of 5×10^{18} electrons/cm^2 (specimen 1) and $\sim 10^{19}$ electrons/cm^2 (specimen 2) and, after that, were annealed for 2 h in an argon atmosphere at a temperature of 800° C. As a pump source, we used the second harmonic of a tunable MaiTai Ti-Sa femtosecond laser radiation (345-520 nm), which was generated by an optical-frequency doubler.

The photoluminescence spectra of specimens 1 and 2 have qualitative similarity. First, their integral photoluminescence intensities do not tend to zero with decreasing pumping wavelength λ_p (as could be expected if one assumes that each of the NV centers has one absorption band in the visible range of the spectrum), but, rather, at $\lambda_p < 390$ nm, they begin to grow. At $\lambda_p > 490$ nm, a "classical" pattern is observed (Fig. 1): against the background of a broad photoluminescence band, narrow peaks

of zero-phonon lines are seen, which indicates that the specimens contain electrically neutral NV^0 and negatively charged NV^- centers (the wavelength of the zero-phonon line is 637 nm for NV^- centers and 575 nm for NV^0 centers). However, as λ_p is decreased, the amplitude of the zero-phonon line of NV^- centers decreases and then the phonon line peak changes into a dip, the amplitude of which increases as λ_p is further decreased.

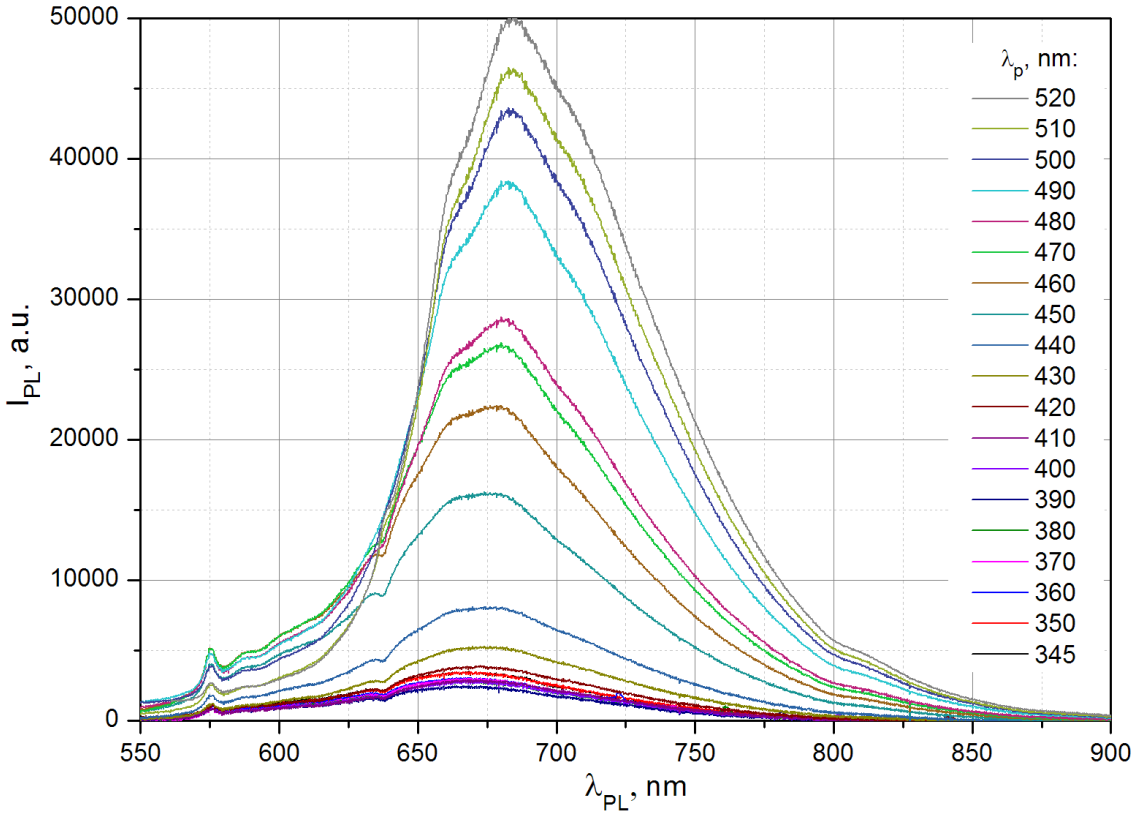

Figure 1. Photoluminescence (Pl) spectra of a diamond crystal with a high content of NV centers (specimen 1). The wavelength of the zero-phonon line is 637 nm for NV^- centers and 575 nm for NV^0 centers. The pumping wavelength λp varies from 520 nm to 345 nm.

We have observed zero-phonon line atop of rather wide ODMR pedestal. The dependence of thus obtained zero-phonon line amplitudes on the pumping wavelength allowed us to conclude that NV^0 centers have an additional absorption line in the UV range [2], which as far as we know, was not observed before. The existence of UV-absorption line was evidenced both by an increase of the NV^0 zero-phonon line amplitude as well as by an increase in the photoluminescence integral intensity, as the pumping wavelength was decreased from 390 to 345 nm. In contrast to NV^0 centers, NV^- centers do not reveal absorption lines in the near UV range. Moreover, as the pumping wavelength is decreased to 495 nm and below, an inversion of the zero phonon line in the luminescence at a wavelength of 637 nm is observed (Fig. 2).

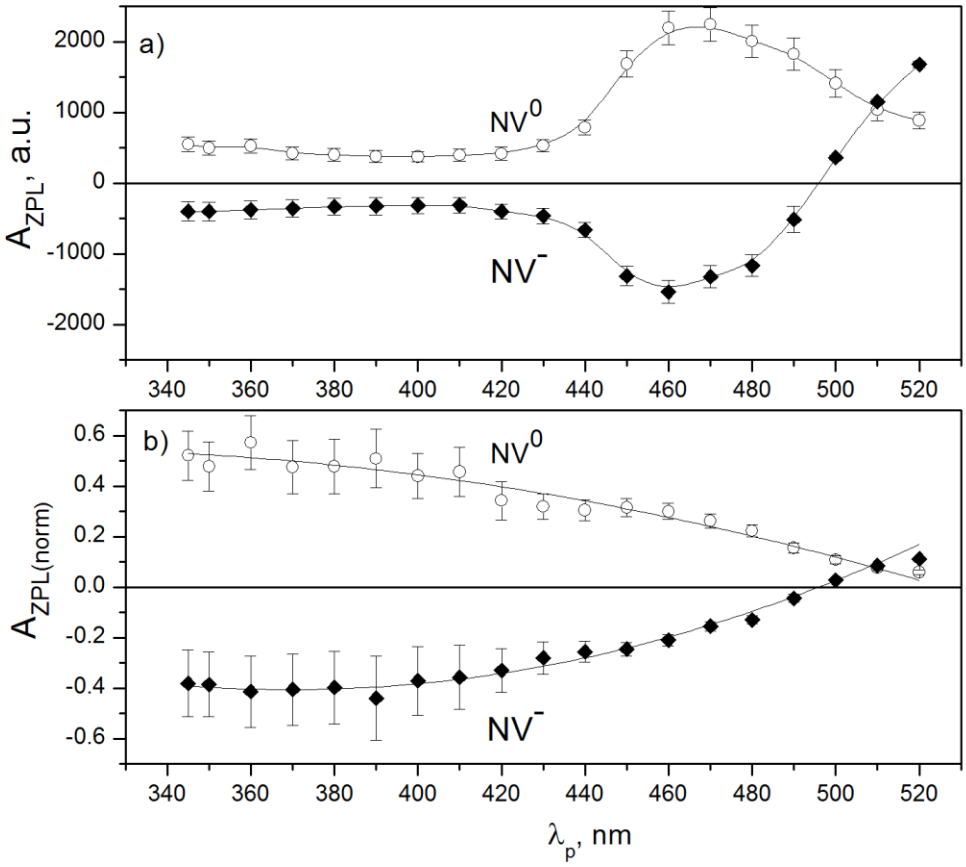

Figure 2. Dependences of the amplitudes of the zero-phonon line (ZPL) of NV^0 and NV^- centers (specimen 1) on the pumping wavelength λp: (a) without normalization and (b) normalization to the average amplitude of the photoluminescence spectrum of the specimen in the spectral interval 550 ÷ 900 nm. Each point corresponds to the line on Fig.1.

This effect can be explained as follows: since typical spectra of NV^- centers are red shifted by 62 nm with respect to the spectra of NV^0 centers [3], the luminescence band of NV^0 centers overlaps both the zero-phonon line of NV^- centers and, partially, the broad phonon absorption band of NV^- centers. Therefore, under the conditions when NV^0 centers absorb the UV pumping radiation and reemit it in the broad band in the range $\lambda \geq 575$ nm, NV^- centers efficiently absorb the light reemitted by NV^0 centers. In particular, this absorption leads to the formation of a narrow dip at the wavelength of the zero-phonon line of NV^- centers (637 nm) in the spectrum of the total photoluminescence of centers of the two types.

3. Observation of ODMR signals in NV^- centers in diamond

ODMR signals were investigated both in high field (as compared with inner fields in the crystal, i.e. above 200 *mkT*), as well as in weak magnetic fields. ODMR resonances were excited with slow

100% amplitude modulation of the microwave field; it allowed us to avoid broadening of the ODMR signal by the modulation frequency.

Frequency splitting of magneto-dependent levels of the ground state of NV^- centers against the magnetic field was measured in the fields (0 ÷ 1) mT and then approximated by formula [3]:

$$2\pi f_{\pm} = [E^2 + (g_s\mu_B B_z)^2]^{1/2} = [E^2 + (\gamma \cdot B_z)^2]^{1/2}, \quad (1)$$

where E – parameter of transverse zero-field splitting, $\gamma = g_s\mu_B$ – gyromagnetic ratio. For the observable sample $E = 2\pi\cdot(3.2 \pm 0.1)MHz$, and gyromagnetic ratio $\gamma = 2\pi\cdot(1.6 \pm 0.1)10^{10}$ Hz/T (Fig.3). Present value is an effective gyromagnetic ratio averaged over all orientations of the crystal. Therefore, according to (1), in typical terrestrial field $B = 5\cdot10^{-5}T$ the maximal achievable sensitivity of a magnetometer based on this sample falls not more than four times compared to the sensitivity achievable in strong fields.

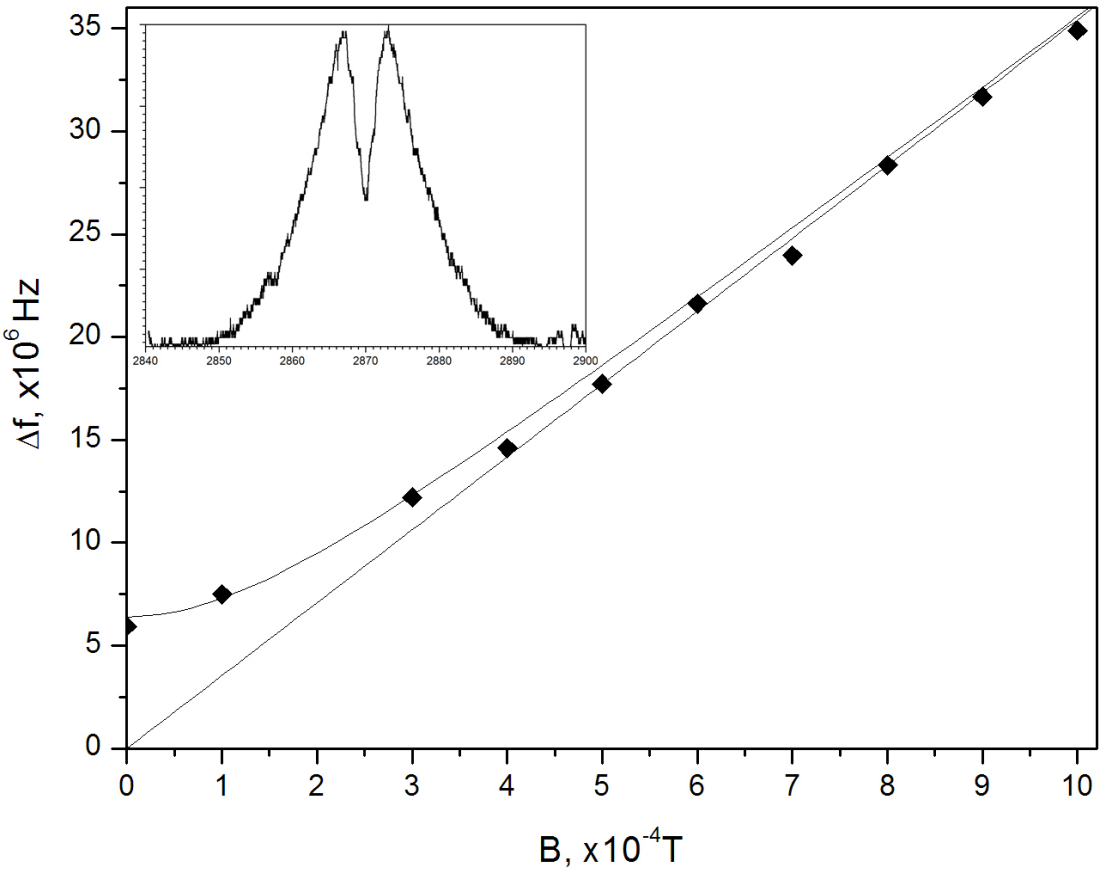

Figure 3. Frequency splitting of magneto-dependent levels of the ground state of NV^- centers against the magnetic field. Inset: zero-field ODMR signal.

Our results prove the feasibility of the sub-millimeter range precision magnetometry of weak magnetic fields ($B \geq 20\ \mu T$) using ODMR in NV^- centers. For this purpose a differential balanced scheme of interrogation of two magnetic microwave transitions ($m_F = 0 \leftrightarrow m_F = -1$ and $m_F = 0 \leftrightarrow$

$m_F = +1$) can be used, which would largely eliminate the temperature dependence of the magnetometer readings. At the same time, the sum of these magnetic transitions frequencies does not carry an information about the magnetic field. Therefore, it can be used for the temperature measurement instead, allowing the final adjustment of the magnetometer readings.

4. Acknowledgments

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