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Diamonds for quantum nano sensing

Taras Plakhotnik

School of Mathematics and Physics, The University of Queensland, 4072 St Lucia, Australia

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ABSTRACT

This paper reviews applications of diamonds for sensing of magnetic and electrical fields, pressure and temperature. Considerable attention is focussed on the interaction of spins with static and oscillating magnetic fields as well as applications of such fields to spin control. A particular focus is on the spin of nitrogen-vacancy centers. Electron-spin microwave resonances play a central role in the ultra-sensitive metrology on the nanoscale, but pure optical methods are also considered in this review.

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1. Introduction

The purpose of this article is to review recent advances in the application of diamonds to quantum sensing on the nanoscale. To start, we need a few definitions clarifying the subject. First, the word *quantum* needs some explanation. It becomes quite fashionable to include *quantum* in a title or at least an abstract or the first paragraph of a paper to make it sound more appealing to readers outside the field. It is believed that the laws of quantum physics (quantum mechanics) form the foundation of all phenomena and therefore as a matter of fact *quantum* can be used almost in any context. But here we will call a measurement *quantum* if the sensitivity of the measurement is limited by quantum fluctuations. A similar motivation exists for the excessively wide usage of the prefix *nano* especially in combination with *technology*. Although any distance can be measured in nanometers, we will use the term *nano* when the characteristic dimensions describing the region of interest for sensing are smaller than approximately 50 nm. The border line is set by the limitations of the resolution in optical imaging due to diffraction.

Application of diamonds for nano-sensing are almost exclusively based on the properties of so-called Nitrogen-Vacancy (NV) centers. Therefore we start with a brief review of the photophysics of these defects in diamond crystals. Such defects have been first described more than 40 years ago [1] but the boom of research began about 20 years later [2]. Then we will briefly review the interaction of electronic and nuclear magnetic moments with micro-wave (MW) radiation and static magnetic fields. The following sections will address applications of nano-diamonds to sensing of magnetic and electrical fields, temperature and pressure. A number of relatively recent review papers are available on various applications of NV-centers [3,4]. These applications are at the proof-of-principle stage and therefore here we will focus more on these principles rather then on the diversity of experimental demonstrations. Examples of applications are included as illustrations in the appropriate sections and in the conclusion.

2. Nitrogen-vacancy centers and spin control

Nitrogen-vacancy (NV) centers in diamond are defects of the crystal lattice (their geometry with C_{3v} symmetry is shown in Fig. 1a) where one out of two neighboring carbon atoms is replaced with nitrogen while the other one is removed without substitution [5]. A single crystal of diamond can host NV centers with four different orientations of their axes. We will refer to such centers as different sets. These sets are not different from each other fundamentally, but the presence of more than one set allows us to define the 3D-vector of a magnetic field relative to these axes. Some of the sets may remain unoccupied in a crystal especially if the total number of centers is small. The wavefunction of the center is localized over a few lattice sites (the reason for calling them *point defects*) and this enables spatial resolution at the *nano* level. Among a large number of known defects in diamond [6] the NV centers are probably most famous. Even though some other crystal defects have been recently proposed to take over the leading role of the NV-centers, the position of the NV-centers has not been shaken. There are two types of such centers – electrically neutral and

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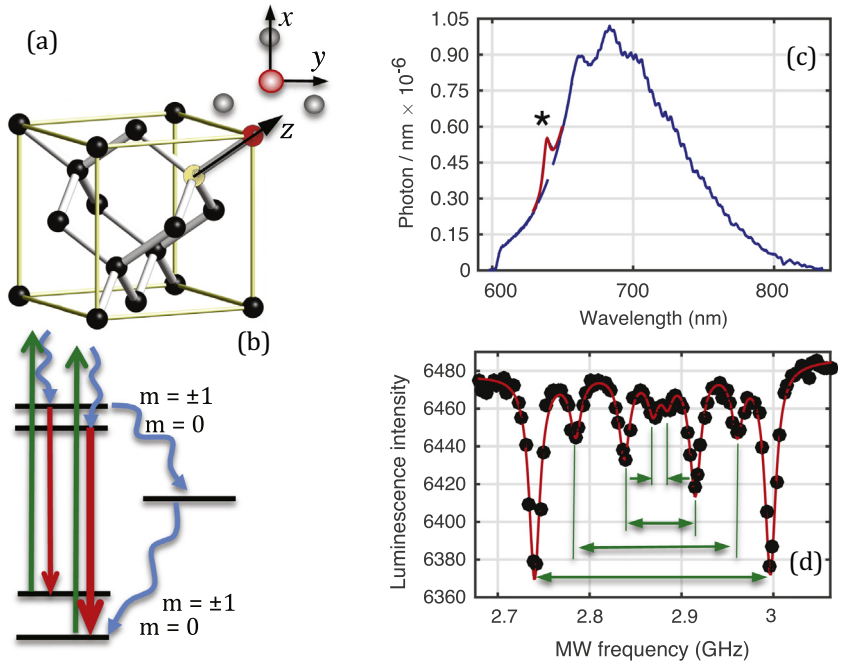

Fig. 1. NV-center in diamond. Panel (a) shows center geometry and the standard orientation of the coordinate system. N and C atoms are in red and black. The vacancy is yellow. The *x*- and *y*-axes are shown as seen from the top of the *z*-axis. Panel (b) depicts the main features of the electronic states at room temperature. Due to double degeneracy of the molecular orbital of the excited spin-triplet state, there are 2 orthogonal transition dipole-moments. Spin-preserving luminescence and optical excitation are shown by the straight arrows downwards and upwards respectively. Panel (c) is an example of a luminescence spectrum (excitation at 590 nm). The ZPL is indicated by an asterisk. The wavy lines indicate non radiative transitions. Panel (d) presents an ODMR spectrum measured (dots) at about 5 mT external field. The 8 Lorentzian-shaped lines (shown by the solid curve) make 4 pairs (indicated by arrows) corresponding to 4 sets of NV centers in the crystal.

negatively charged. We will focus on NV^-. Their unique properties which are most important for sensing applications are strong and resistant to bleaching photo luminescence with a zero-phonon line (ZPL) that is prominent even at room temperature, electron spin-triplet nature of the electronic ground state (see Fig. 1b), and strong dependence of the luminescence intensity on the value of the spin projection on the symmetry axis of the center. An example of the luminescence spectrum of NV-centers is shown in Fig. 1c. A small feature at about 638 nm is the ZPL. The ZPL results from optical transitions which do not change the phonon population in the crystal. The rest of the spectrum is called *phonon wing* and is made by photons which were generated during the electronic energy relaxation accompanied by creation or annihilation of one or several phonons. Due to the dependence of the intersystem crossing on the magnetic quantum number m, $(m = 0, \pm 1)$, the luminescence is strong when the center is in the $m = 0$ state and is weak when the populated state is either of $m = \pm 1$. When the center is unperturbed, the energies of the $m = \pm 1$ states are nearly equal and about 2.87 GHz (the units of frequency are obtained when the energy is divided by the Planck constant and will be frequently used here instead of joule) higher than the energy of the $m = 0$ state. At room temperature, the energy difference between the spin states is much smaller than the thermal energy k_BT (the product of the Boltzmann constant and the temperature) and therefore the probabilities that a particular NV center occupies one of the three spin sub-levels are practically equal. But optical pumping in an about 100-nm broad band centered near 575 nm wavelength transfers most of the population to the $m = 0$ level. This phenomenon is called spin polarization by optical pumping or simply optical spin-polarization. The reason for such polarization is again a much higher probability of the intersystem crossing from the triplet $m = \pm 1$ levels to singlet states sitting between the triplet ground and the first excited triplet states followed by the relaxation to the electronic ground $m = 0$ state. Such a relaxation path effectively converts population of the $m = \pm 1$ states into population of the $m = 0$ state. Resonance MW-radiation at 2.87 GHz repopulates the $m = \pm 1$ states and this can be observed optically as a decrease in the luminescence intensity. Such a phenomenon enables optical detection of magnetic resonance, abbreviated as ODMR. An example of an ODMR spectrum is shown in Fig. 1d. The Hamiltonian of an NV-center in the presence of external magnetic and electrical fields reads

$$\widehat{H} = \mathcal{D}\widehat{S}_z^2 + \mathcal{E}_x(\widehat{S}_x^2 - \widehat{S}_y^2) + \mathcal{E}_y(\widehat{S}_x\widehat{S}_y + \widehat{S}_y\widehat{S}_x) + \gamma_e\mathbf{B}\widehat{\mathbf{S}} \qquad (1)$$

In this equation, the coordinate system is chosen so that its *z*-direction coincides with the direction of the NV axis and the *x*-axis is positioned in the plane going through N, V and one of the nearest C (see Fig. 1a). Dimensionless ($\hbar$ not included) spin 1 operators $\widehat{S}_{x,y,z}$ are used for briefness. If $\mathbf{B} = 0$ and $\mathcal{E}_{x,y} = 0$, the value of $\mathcal{D} \approx 2.87$ GHz determines the zero-field splitting between $m = 0$ and the degenerate $m = \pm 1$ levels. The values of $\mathcal{E}_x = \mathcal{E}_\perp \cos\phi_\mathcal{E}$ and $\mathcal{E}_y = \mathcal{E}_\perp \sin\phi_\mathcal{E}$ characterize the stress in the crystal perpendicular to the symmetry axis of the NV center ($\phi_\mathcal{E}$ is the polar angle in the *xy* plane). Such stress mixes the $m = \pm 1$ states into $|\pm\rangle \equiv \frac{1}{\sqrt{2}}\left(\exp\frac{i\phi_\mathcal{E}}{2}|1\rangle \pm \exp\frac{-i\phi_\mathcal{E}}{2}|-1\rangle\right)$ which have energy split of $2\mathcal{E}_\perp$. Note that even in nano-crystals where the stress is typically much larger than in a nearly perfect bulk diamond, the numerical value of $\mathcal{E}_\perp$ is about three orders of magnitude smaller than the value of $\mathcal{D}$. The coordinate system can be rotated around *z* axis so that the expression $\mathcal{E}_x(\widehat{S}_x^2 - \widehat{S}_y^2) + \mathcal{E}_y(\widehat{S}_x\widehat{S}_y + \widehat{S}_y\widehat{S}_x)$ is reduced to $\mathcal{E}_\perp(\widehat{S}_x^2 - \widehat{S}_y^2)$. Such a choice simplifies the Hamiltonian (1) but may be inconvenient for treatment of, for example, spin-lattice coupling.

It is apparent from Eq. (1) that the position of the EPR resonances will depend on the strength of the external magnetic filed **B** which will affect mainly the energies of the $m = \pm 1$ levels. This sensitivity can be used to measure the strength and direction of

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