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Measurement of single electron and nuclear spin states based on optically detected magnetic resonance

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Abstract. A novel approach for measurement of single electron and nuclear spin states is suggested. Our approach is based on optically detected magnetic resonance in a nano-probe located at the apex of an AFM tip. The method provides single electron spin sensitivity with nano-scale spatial resolution.

1. Introduction

Progress in nanotechnology, including spintronics and quantum information processing based on a solid-state quantum computer, has brought significant attention to the problem of measurements of single electron and nuclear spin states. Here we suggest a novel approach for measurement of single electron and nuclear spin states. The novel aspects of our approach are: (1) use a nano-probe for the optically detected magnetic resonance (ODMR), (2) non-destruction of the spin state being measured, (3) nano-scale spatial resolution, and (4) high sensitivity of ODMR to a single electron or nuclear spin orientation relative to an external magnetic field. Single electron spin measurements in molecules were independently performed in 1993 by two groups led by Moerner [1] and Orrit [2]. Today, the principles of detection of a single electron spin based on ODMR are well established. The limitation of the lateral resolution of ODMR is related to the size of the light spot. The highest resolution is obtained by a near-field scanning optical microscope, which has light spot size of about 30-50 nm. Another limitation of the ODMR technique is that the unpaired electron has to be a part of a molecule, which absorbs or emits light. Our modified ODMR approach consists of an AFM with a photoluminescent material (nanoparticle) located at the apex of the AFM tip (see Fig. 1), which exhibits ODMR in the vicinity of an unpaired electron or nuclear spin in the sample. This approach transfers the detection of electron magnetic resonance from a microwave frequency domain to an optical domain that significantly increases measurement sensitivity.

In a semiconductor, quantum-dot confinement leads to a replacement of continuous bands of energy by molecular-like energy level structures. The theory of spectrum of quantum dots shows that the ODMR spectrum depends on the mutual hole-electron interaction in the exciton. In the ODMR method, the nanoparticle is excited from its ground state 1 into the first excited state 2 by absorption from a laser field, and subsequently decays through non-radiative transitions to magnetic sublevels states 3 (See Fig. 2). An

external magnetic field lifts the degeneracy of the triplet states and changes the energy splitting between these magnetic sublevels as well as their population and lifetimes. Resonance is achieved by scanning the magnetic field from a nearby radio-frequency coil to induce transitions between these magnetic sublevels to change their relative populations, thus increasing or decreasing the intensity of specific peaks in the photoluminescence spectrum resulting from decay back to the ground state.

The proposed optical illumination scheme known as an apertureless near-field scanning microscopy probe leads to significant enhancement of optical field in the vicinity of a sharp tip. The intensity enhancement near a silicon tip can be of several orders of magnitude. This enhancement arises from the antenna effect of the elongated probe. Thus nanoprobe absorption in an evanescent laser field could be significantly enhanced when the probe is placed at the apex of a sharp silicon tip. The potential resolution of this method is related to the size of the photoluminescent probe, typically 1 - 10 nm. There are two possible types of measurements. In the first type of measurement, the frequency of the oscillating magnetic field is fixed and the external permanent magnetic field is varied. In the second type of measurements, the permanent magnetic field is fixed and the frequency of oscillating magnetic field is varied in the vicinity of the resonance frequency. One of the most promising applications for this method is the non-destructive measurement of a quantum bit (qubit) single spin state in a quantum computer (QC).

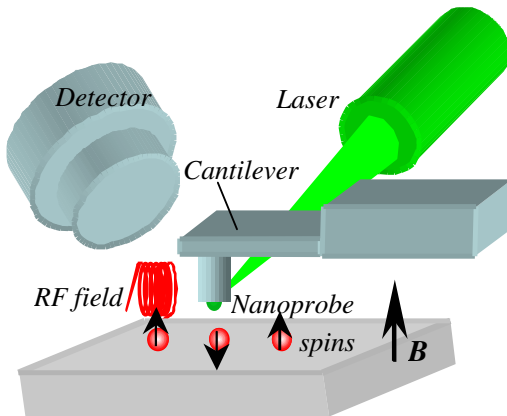


Figure 1. AFM-ODMR setup.

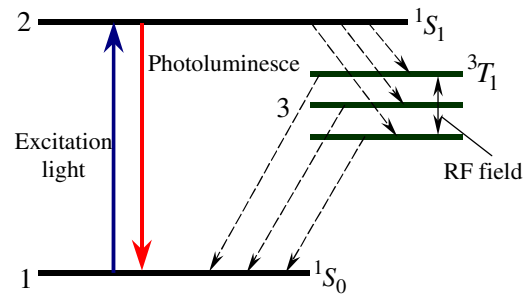


Figure 2. Transitions in the nanoparticle.

2. Measurement of a single electron spin state

The preliminary analysis of the dependence of the sensitivity on geometry shows that at optimal conditions the ODMR of a nano-size probe can sense the magnetic field of a single electron spin [3]. Experiments with a variable magnetic field demonstrate rather sharp spectral structures in the range of 0.1 – 0.002 T at a sample temperature of about 4 K [4, 5]. To estimate the sensitivity limit of an ODMR-based scanning microscope, we assume that the diameter of the nanoprobe is 1 nm, the distance between the nanoprobe surface and the sample is 5 Å, and within a radius of several nm there is only one unpaired spin. The magnetic field from a single spin is given by $\mathbf{B}_s = (\mu_0/4\pi)(3\mathbf{n}(\mathbf{m})-\mathbf{m})/r^3$. When we substitute $\mathbf{m} = -(1/2)g_e \mu_b = -9.28 \times 10^{-24}$ J/T, $\mu_0/4\pi = 10^{-7}$ N/A², we get $B_s = 1.5 \times 10^{-2}$ T. This value of magnetic field of an individual spin is larger than the most narrow features of the ODMR spectrum, and could be detected by measurement of the resonance shift.

It is very desirable that the procedure for measurement of the static magnetic field of a single spin does not perturb its quantum state and, therefore, could be considered a non-destructive measurement of the quantum object. This property depends strongly on the quantum nature of exciton in the nanoprobe. For example, an exciton transfer from the nanoprobe to the impurity atom with unpaired electron spin or a vice versa process can change the initial electron spin state. These kinds of processes are forbidden if there are no coincidences of exciton energies in the nanoprobe and in the impurity atom. The energies of magnetic sublevels in the strong external magnetic field in the nanoprobe and in the impurity atoms usually are different because the difference in the symmetry and the value of the fine and hyperfine interactions. In opposite to the case of the impurity atoms, in the nanoprobe the splitting of energy of magnetic sublevels occurs even at the absence of the external magnetic field. The detailed consideration of these processes lies outside the scope of the present article, and will be a subject of further study.

3. Measurement of a single nuclear spin state

We consider the two states of a single nuclear spin of an impurity atom with an electron inside a solid-state matrix (Fig. 3). Fig. 4 presents the scheme of energy levels of electron-nuclear spin states in the presence of a permanent magnetic field. The energy levels of an electron spin are shifted due to the hyperfine interaction, which depends on the state of the nuclear spin. To measure the state of the nuclear spin, we exploit the fact that the ESR resonances for transition frequencies between sub-levels of the electron spin depend on the state of the nuclear spin (see Fig. 4). Thus, for example, the RF field will selectively induce transitions only between the states $\langle S_e = -1/2, I_n = 1/2 \rangle$ and $\langle S_e = 1/2, I_n = 1/2 \rangle$, and will not interact with transition $\langle S_e = -1/2, I_n = -1/2 \rangle - \langle S_e = 1/2, I_n = -1/2 \rangle$. Finally, the two electron spin states involved in the transitions correspond to the same nuclear spin “up” state. Therefore, the proposed measurement procedure does not change the state of a nuclear spin, and realizes a non-destructive measurement of a nuclear spin state.

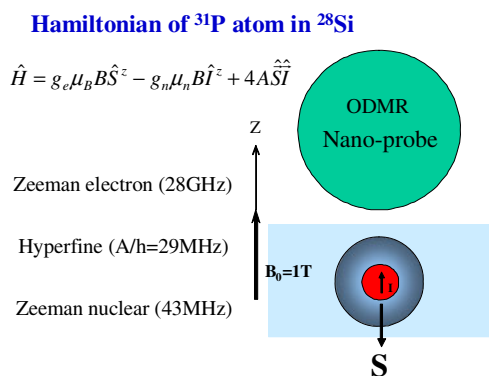


Figure 3. Nanoprobe and ^{31}P atom in ^{28}Si matrix. Relevant energies are shown at left.

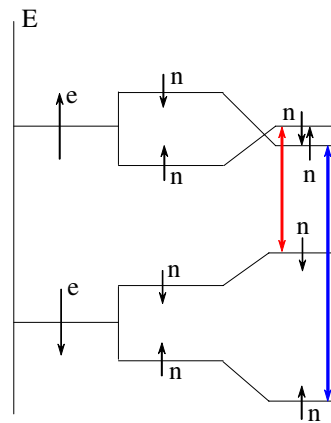


Figure 4. Hyperfine structure of electron-nuclear spin energy levels.

4. Approach to developing functionalized AFM optical sensor probes

We consider two types of promising candidate sensor materials that exhibit ODMR properties: 1) nitrogen-vacancy (N-V) center in diamond-like carbon (DLC) [4] and 2) CdSe nanoparticles [5]. Each

type of material is optically active, has reasonably sharp photoluminescence peaks, and has optical properties that are sensitive to magnetic fields. The most straightforward way to develop a probe with a DLC tip at the apex of an AFM tip is to grow a fiber directly on the tip in a scanning electron microscopy. This method will allow reasonably rapid production of sharp carbon tips with N-V centers created by the electron bombardment. CdSe quantum dots that are 2-10 nm in diameter are also expected to exhibit sharp ODMR signals [5] that could provide the resolution needed to detect the influence of a single spin in very close proximity. One of the challenges of application of CdSe quantum dots is to synthesize CdSe/ZnS or CdSe/CdS core/shell quantum dots of high quality and with as thin a ligand shell as possible. The quality as well as the wide-band gap shell material are needed to minimize blinking and spectral diffusion, and also to avoid impurities such as oxygen vacancies that would add additional features to the ODMR signal. In addition, one would like to have good control over the QD size and size dispersion.

5. Conclusion

We propose a novel approach for non-destruction measurement of a single electron or nuclear spin state. Our approach is based on OMDR in a nanoprobe located at the apex of an AFM tip, and provides a nanoscale spatial resolution.

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