Current Applied Physics 14 (2014) 1234–1239

Contents lists available at ScienceDirect

Current Applied Physics

journal homepage: www.elsevier.com/locate/cap

Optically controlled initialization and read-out of an electron spin bound to a fluorine donor in ZnSe

Y.M. Kim [a, d, *], D. Sleiter [b], K. Sanaka [b, c], D. Reuter [a], K. Lischka [a], Y. Yamamoto [b, c], A. Pawlis [a, b]

[a] Department Physik, Universität Paderborn, Warburger Str. 100, 33098 Paderborn, Germany
[b] Edward L. Ginzton Laboratory, Stanford University, Stanford, CA 94305-4088, United States
[c] National Institute of Informatics, 2-1-2 Hitotsubashi, Chiyoda-ku, Tokyo 101-8430, Japan
[d] Korea Advanced Institute of Science and Technology, 291 Daehak-ro, Yuseong-gu, Daejon 305-701, Republic of Korea

ARTICLE INFO

Article history:
Received 4 April 2014
Received in revised form
5 June 2014
Accepted 17 June 2014
Available online 2 July 2014

Keywords:
Electron spin
Qubit
II–VI semiconductor
Nanostructures
Initialization
Read-out

ABSTRACT

Here we report photon antibunching and magneto-spectroscopy of a single electron spin bound to a fluorine donor in a ZnMgSe/ZnSe QW nanostructure. The results confirm the presence of an optically controllable lambda-system which allows the optical manipulation of the electron bound to the neutral fluorine donor as a spin qubit. Moreover, we achieved optical spin pumping of the qubit by resonant excitation of each of the four allowed transitions of the lambda system. We verified the spin transfer by detecting single photons when the bound electron decays into the opposite spin state. The results presented here constitutes an elegant initialization and the read-out procedure of the electron spin qubit bound to a fluorine donor which are prerequisite for coherent optical control of an impurity based solid-state spin qubit.

1. Introduction

Many proposed schemes for quantum information processing require two fundamental qubits properties: homogeneity and scalability [1–5]. In different qubits systems, it has been reported that a system which possesses an inherent advantage of one, in general shows a need to be improved on the other property: Trapped ion qubits are extremely homogeneous and have nearly perfect optical quantum efficiency [6–8] but scalability is inherently difficult due to the need of trapping and cooling of the ions to extremely low temperatures. Also, in a large set of ions, they become more susceptible to noisy electric field, and decoherence of the motional modes could also occur [9]. On the other hand, semiconductor based qubits such as self-assembled quantum-dots (QDs) possess superior scalability, but homogeneity is more critical largely due to the randomness in dot formation size [10–13]. Since most of the qubit applications demonstrated with QDs are based on III–V compound semiconductors (e.g. GaAs, InAs and alloys) a further limitation is the inevitable presence of non-zero nuclear spins in this material system which results in a reduced decoherence time up to few microseconds [14]. The latter can be enhanced by dynamic decoupling [15] but this requires applying complex pulse sequences, which reduce the speed of qubit manipulation. Other qubit implementations such as charged nitrogen-vacancy (NV) centers in diamond [16], or electron spins bound to shallow impurities in semiconductors [17] are particularly suited to bridge the gap between excellent homogeneity and scalability. For instance, charged NV centers in diamond are more homogeneous than quantum dots, and yet they retain decent properties [18,19]. The low optical quantum efficiency of this system, i.e. about 3% of their emission in the zero-phonon line, demands for high-quality cavities for enhanced collection efficiency [20]. Moreover, like the QDs based on III–V system, one of the major sources of decoherence of the NV electron spin is the interaction with the nuclear spins resulting in a typical decoherence time of few μs. This can be suppressed by dynamical coupling technique augmenting the decoherence time to 10 ms regime, although it reduces the speed of the entanglement protocol in exchange [21]. Remarkably, the entanglement between two isolated NV-center electron spin qubits

* Corresponding author. Korea Advanced Institute of Science and Technology, 291 Daehak-ro, Yuseong-gu, Daejon 305-701, Republic of Korea.
E-mail address: ymartinkim@kaist.ac.kr (Y.M. Kim).

http://dx.doi.org/10.1016/j.cap.2014.06.017
1567-1739/

over a distance of three meters applying this technique was demonstrated in a recent work [21]. Alternatively, electron spins bound to individual donors in suitable direct-bandgap semiconductors provide high oscillator strength and large exciton binding energies. As a matter of fact, those donors form atom-like transitions with high-optical quantum efficiency and in addition constitute a scalable system [22–27].

Here we review recent key achievements by making use of a single electron trapped by a fluorine donor in ZnSe as a suitable electron spin qubit [23,25,26]. It forms an optical lambda system in a magnetic field similar to that used in QD qubits, which allows for manipulation and coherent control of the spin state of the electron by optical pulses. The optical quantum efficiency of the donor system is very close to unity [27] and can be used to generate triggered single photons [24]. Furthermore, it has recently been shown that photons from two independent nanostructures, each containing an isolated fluorine atom, can be indistinguishable [24] and polarization-entangled through post-selection [28]. While coupling between the electron spin and the nuclear spin of the host crystal is the leading decoherence-causing mechanism in III–V semiconductor QDs [29], isotopic purification can be used to deplete the ZnSe host crystal of the low amount of remaining non-zero nuclear spins [17]. This is not possible in III–V semiconductors but has been very successful to extend the coherence times of electrons in diamond and silicon [30,31]. Furthermore, fluorine has a natural 100% abundance of spin-1/2 nuclei, which may be proposed as an additional spin qubit naturally coupled to the electron spin qubit [26,23,32,33] via hyperfine interaction. Optically active fluorine donors have also been successfully implanted in ZnSe using ion implantation, which could eventually lead to deterministically placing single donors in specific locations [25]. Recently, it has been shown that an ensemble of fluorine donors in bulk ZnSe features long electron-spin dephasing times T_2^*, exceeding 30 ns for temperatures up to 40 K [34]. While, the presence of the inhomogeneous broadening likely introduced by strain and surface imperfection in nanostructures could reduce the homogeneity of this system, it possesses superior inhomogeneous to homogeneous linewidth ratio to that of the III–V QDs, and the NV-centers in diamonds [26,20]. All of these characteristics make the ZnSe:F system an appealing qubit candidate for application in solid-state quantum information processing schemes.

The initialization and read-out of single electron or hole spin in the QDs have been both theoretically [35] and experimentally [36] reported. The first demonstration of the optical pumping of a single electron bound to a fluorine donor (in the following denoted fluorine qubit) in a ZnMgSe/ZnSe QW nanostructure was reported in Ref. [26] where mainly one optical transition was analyzed. Here we provide comprehensive comparison of all four individual possible pumping and detection combinations as well as more detailed review of the magneto-optical properties. Both, the previous result together with the detailed study presented in this article provide substantial evidences for the initialization and the read-out as well as the fully-connected nature of our optical lambda system.

2. Material and methods

The ability to initialize qubits to a known state, and to read-out of their final state are important prerequisites in quantum information processing technology [1,2,5]. Here, we first describe the relevant optical transition-level scheme of an isolated fluorine-bound electron-spin within ZnMgSe/ZnSe QW. Subsequently, we present experimental evidence of the proposed optically controllable system under an external magnetic field. Photon correlation measurements were carried out to confirm the isolation of a single fluorine qubit by the radiative recombination of excitons bound to this donor. Finally, we focus on the optical pumping experiments that we have performed on all four branches of the lambda-system and discuss the results that clearly indicate successful initialization and read out of the spin state of the fluorine qubit. Fig. 1a shows a typical sample structure of fluorine δ-doped ZnMgSe/ZnSe QWs. The ZnMgSe/ZnSe/ZnMgSe quantum structure is grown by molecular beam epitaxy (MBE) on an about 20 nm thick ZnSe buffer-layer on top of GaAs substrates. The first ZnSe layer directly on GaAs substrate serves to avoid abrupt interface between two chemically unalike materials for superior optical quality. The ZnSe QW layer has a thickness of 2 nm and is enclosed between two 28 nm thick ZnMgSe cladding layers with a magnesium content of about 17%. Furthermore, the ZnSe QW is fluorine δ-doped in its central monolayer. The δ-doping is applied to reduce the spectral distribution of the relevant transitions [23] by localizing the fluorine donor in the center of the QW, and to avoid forming of p-like states of donor-bound excitons close to the interfaces of the QW. For the isolation of single fluorine donors, we defined nano-pillar structures with 100–200 nm diameter by e-beam lithography and subsequent wet-chemical etching by Potassium dichromate solution ($K_2Cr_2O_7$). By considering a typical sheet δ-doping concentration of approximately $1–3 \times 10^{10}$ cm^{-2}, about 1–4 fluorine atoms can be expected within each nanostructure.

When a fluorine impurity replaces a Se atom in ZnSe, it forms a shallow donor potential with ionization energy of about 29 meV in bulk, [37]. This is termed as the neutral donor state (D^0) assigned in Fig. 1b. At sufficiently low temperature, e.g. <50 K, the fluorine impurity can capture a free exciton forming a donor bound exciton complex (D^0X) with binding energies exceeding 10 meV due to additional confinement in a ZnMgSe/ZnSe QW nanostructure. The $D^0X \rightarrow D^0$ transition refers to the radiative recombination of this bound exciton by the emission of a single photon. Here we used a 2 nm thick ZnSe QW with a $D^0X \rightarrow D^0$ transition energy of 2.865 eV [34,38]. When an external magnetic field is applied, both the D^0 and D^0X state split via the Zeeman Effect as shown in Fig. 1b. The two non-degenerate spin states in D^0 form the two qubit basis; $|0\rangle$ and $|1\rangle$, which can be optically connected to $|e\rangle$ of the D^0X state, forming an optical lambda-system. The qubit state of D^0 is thus defined by these two non-degenerate bound electron spin states, while that of the D^0X state is determined by the heavy-hole, since the two electrons in D^0X form a singlet. Fig. 1c and d depicts the two magnetic geometries of choice; Faraday and Voigt respectively. In Faraday geometry, the magnetic field, $\vec{B}$ is parallel to the QW-confinement direction, $\vec{z}$, while in Voigt, the magnetic field, $\vec{B}$ is perpendicular to the QW-confinement direction, $\vec{z}$. The allowed optical transitions in both geometries are significantly different: In Faraday, the spins of both the electron in D^0 and the heavy-hole in D^0X are aligned together in z-direction, and following the selection rules, two circular polarizations; σ^+, σ^- are determined as shown in Fig. 1c. As a result, there is no allowed optical transition connecting the electron spin ground states $|0\rangle$ and $|1\rangle$ in Faraday geometry. On the other hand, in Voigt geometry, the spin states of the electron and heavy-hole are aligned differently: the electron in D^0X has s-like orbital wavefunction which is spherically symmetric; thus its spin can be aligned according to the direction of the applied magnetic field. The heavy-hole in D^0X however, has p-like orbital wavefunction which is not spherically symmetric; thus, the heavy-hole spin is dominantly aligned by the QW-confinement in the z-direction. As a result, the applied magnetic field acts as a mere perturbation to the dominant QW-potential, causing the heavy-hole spin states to be mixed; thus the heavy-hole spin in the D^0X forms a superposition state of $|+3/2\rangle$ and $|-3/2\rangle$. The latter effect introduces two additional transitions, cross-connecting the ground states $|0\rangle$ and $|1\rangle$ of the electron spin qubit via the corresponding