Photon Antibunching and Magnetospectroscopy of a Single Fluorine Donor in ZnSe

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We report on the optical investigation of single electron spins bound to fluorine donor impurities in ZnSe. Measurements of photon antibunching establish the presence of single, isolated optical emitters, and magneto-optical studies are consistent with the presence of an exciton bound to the spin-impurity complex. The isolation of this single donor-bound exciton complex and its potential homogeneity offer promising prospects for a scalable semiconductor qubit with an optical interface.

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Schemes for quantum information processing and quantum communications rely on scalable, robust qubits. In particular, there are many proposals that require fast, efficient, and homogenous single-photon sources[1–3] and still others that rely on the interaction between matter qubits and flying photonic qubits[4]. The requisites for both types of schemes can be satisfied with semiconductor electron spins, which serve as single photon sources[5] or long lived quantum memories with an optical interface[6–8]. However, optical schemes, particularly those based on entanglement, also require large numbers of homogenous photon emitters[9–14]. Electron spins in self-assembled QDs, unfortunately, suffer from large inhomogenities due to their natural size distribution.

Impurity-bound electrons in direct bandgap semiconductors, however, have relatively little inhomogeneous broadening[15–20], yet still possess strong optical transitions when binding an additional exciton[18–21] and long ground state coherence times[7].

An electron bound to a single fluorine donor in ZnSe (F:ZnSe) may serve as a physical qubit with many potential advantages over previously researched qubits. F:ZnSe is particularly appealing because of its nuclear structure compared to III-V-based bound-exciton or quantum dot systems. Unlike III-V systems, isotopic purification of the ZnSe-host matrix to a nuclear-spin-0 background is possible, eliminating magnetic noise from nuclear spin diffusion[22,23]. Further, the F-impurity has a nuclear spin of 1/2 with 100% abundance. Electron-nuclear spin swapping schemes[24,25] can be used, which, in combination with the spin-0 background of the isotopically purified host matrix, could lead to an extremely long-lived qubit. Additionally, the applicability of standard microfabrication techniques[26,27] to ZnSe makes the F:ZnSe system particularly scalable.

The F:ZnSe system has already shown promise as a scalable source of single photons in Ref. 20. However, this work did not demonstrate the potential of the donor system as a future quantum memory. Here, we show both statistics for single photon emission, as well as the presence of a three-level optical Λ-system through magnetospectroscopy experiments. This introduces F:ZnSe as a valid candidate for use as a scalable qubit with an optical interface.

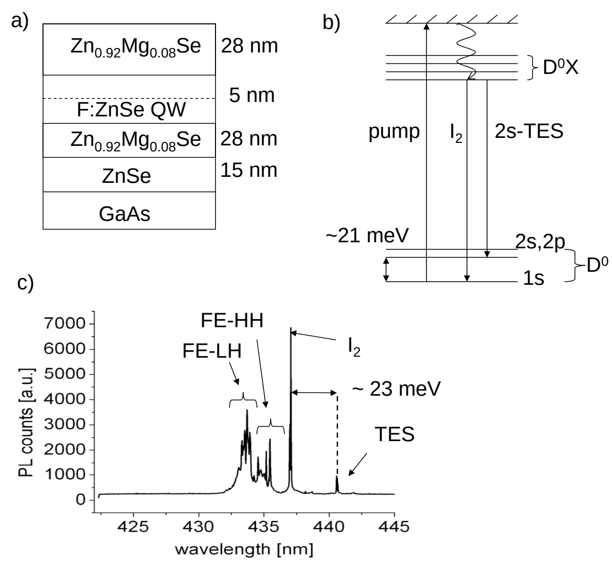

FIG. 1. Sample structure and PL data. a) Schematic of sample structure: the dotted line indicates the location of the δ-doping. b) level structure: D^0 represents the F-bound-electron manifold with its excited electron states, while D^0X represents the F-bound exciton manifold. Excitation occurs through above-band pumping, with fast non-radiative relaxation into the bound-exciton ground states. Radiative decay occurs into either the ground state (indicated by I_2) or excited states (TES). c) mesa-structure (100 nm diameter) PL. FE-HH and FE-LH represent the heavy and light hole free exciton emission, while I_2 and TES indicate the bound exciton decay into the respective electron ground and excited states.

The sample structure is illustrated in Fig. 1(a). The samples that were studied all consist of an MBE-grown,

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fluorine δ-doped ZnSe quantum well (QW), ranging between 1 and 10 nm in thickness, sandwiched between two ZnMgSe cladding layers (Mg content approximately 8%). GaAs-(001) substrates were used, with a thin, undoped ZnSe buffer layer defining a clean interface for the II-VI/III-V heteropitaxy. The ZnMgSe cladding layers prevent carrier diffusion into the lower-bandgap GaAs substrate and spectrally shift the background ZnMgSe emission from the F:ZnSe photoluminescence (PL). The areal Fluorine impurity δ-doping density was approximately 8 x 10^9 cm^{-2}. δ-doping was chosen both to ease the isolation of individual impurities by reducing the total number of dopants in the QW, as well as to locate the dopant impurities in the center of the QW. Spatial isolation of individual emitters occurred through mesa-etching, where selected parts of the Zn(Mg)Se-sandwich layer were chemically removed in a $K_2Cr_2O_7$:HBr:H_2O (1:130:250) etch mixture[28]. The nominal diameter of the mesas varied from 50 to 400 nm, and the mesas were capped in SiO_2 for chemical stability.

PL characterization was performed in a cold-finger, liquid-He cryostat at 10 K, using a 408 nm laser diode acting as above-band pump source, similar to the experiments described in Refs. 29,30. The relevant transitions are schematically illustrated in Fig. 1(b). A spectrum is shown in Fig. 1(c). Note that several peaks are visible: I_2 and the two-electron satellites (TES)[19] are related to the bound exciton decay, whereas FE-HH and FE-LH represent a continuum of heavy- and light hole free-exciton emission respectively. In contrast to the decay into the ground state (I_2), the TES are associated with decay into excited states of the bound electron. From a zeroth-order Bohr-model, these are expected to be redshifted from I_2 by about 21 meV, which is close to the experimental value of 23 meV, hence establishing the origin of the emission peaks as related to donor-bound exciton transitions. From separate, F-concentration dependent growth studies (not shown), we infer its origin as F-impurity bound exciton decay.

In order to verify whether a single peak corresponds to a single emitter, photon correlation measurements of $g^{(2)}(\tau)$ were performed[31]. In these experiments, an aboveband pulsed laser excites the mesa of interest and the emitted photons are frequency-filtered using a grating and a slit. This filtered light is sent to a beamsplitter with a detector at each output arm. Coincidence clicks between the detectors are counted as a function of the delay, τ, between those clicks. After normalization by single detector clicks, the count rate, $g^{(2)}(\tau)$ gives insight to the nature of the light source via its photon statistics. In such a measurement the correlation function $g^{(2)}(\tau)$ for a perfect single emitter without a background approaches zero at zero delay ($\tau = 0$)[5,32]. A mode-locked laser with repetition rate of 76 MHz was used for above-band pumping, and silicon avalanche-photodiode-based single-photon-counting modules were used with a time-interval analyzer for timing photon detection events.

The lowest $g^{(2)}(0)$ measured to date in these samples is

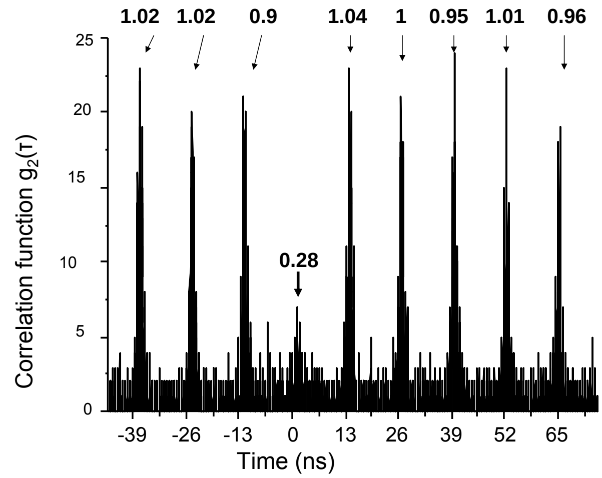

FIG. 2. Non-normalized $g^{(2)}(\tau)$ correlation function measurement for a 100 nm mesa. Normalized peak values (1 ns integration window around the peaks) are indicated above the peaks . Note the $g^{(2)}(0)$-dip, denoting antibunching, and the repetition rate of 13 ns or 76 MHz.

0.28, as shown in Fig. 2 for a mesa structure with diameter 100 nm, QW-thickness of 5 nm. This value confirms the presence of an individual emitter, since it is less than the threshold value of 0.5 that excludes two or more photons being emitted[32]. The origin of the 0.28 background is attributed to the presence of nearby emitters, especially from the tail of the free-exciton emission peak.

While the measurement of $g^{(2)}(0)$ establishes F:ZnSe as a viable single-photon source, its use as an optically controllable qubit requires verification of the presence of a three-level Λ-system. For this reason, magnetospectroscopy measurements were performed in a 10 T superconducting magnet at 1.5 K. A room-temperature reentrant bore window allowed for a microscope objective (working distance: 38 mm, NA: 0.18) to be used for collection of the photoluminescence. Above-band illumination from a CW, 408 nm GaN laser-diode was chosen for the excitation of the sample, and the photoluminescence spectrum was collected through a 750 mm spectrometer on a liquid-N_2-cooled CCD-camera. Both Faraday and Voigt geometry data were obtained through the use of small mirrors inside the cryostat (We refer to the insets of Fig. 3(b) and (d) for the respective orientations).

The expected energy spectrum as a function of applied magnetic field is shown in Figs. 3(a) and (c) (See also Refs. 18, 21, 33, 34 and 35 for a review on magnetospectroscopy of bound-exciton systems). Note that, due to the QW and compressive strain in the ZnSe-layer, the degeneracy between the heavy and light holes is lifted[29,30].

In Faraday geometry, a two-fold split of the photoluminescence is observed. This is consistent with the model of an exciton bound to an electron-impurity complex. From the linesplit, we infer a difference in the out-of-plane heavy-hole and electron g-factors ($\mid 3g^{\parallel}_{\mathrm{hh}} - g_{\mathrm{e}} \mid$) of 0.8 ($\pm$0.2).

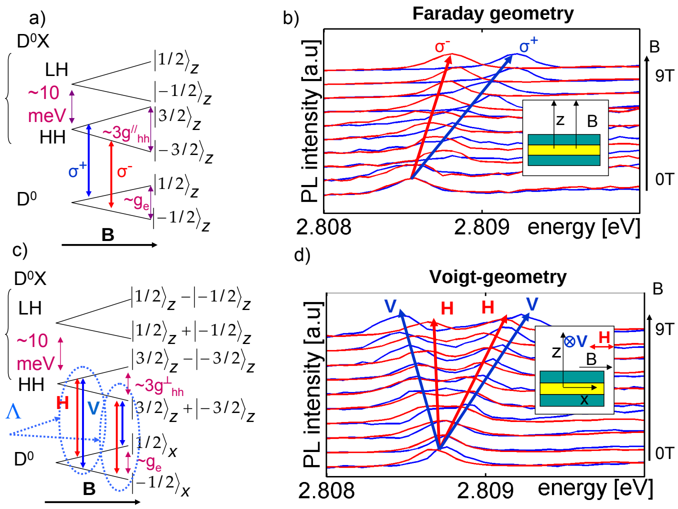

FIG. 3. Magnetospectroscopy data for 200 nm diameter mesas. a) Energy spectrum in Faraday geometry (D^0X: bound exciton, D^0: bound electron, HH: heavy hole, LH: light hole); b) PL as function of magnetic field, in Faraday geometry (inset: sample and field alignment in Faraday geometry); c) Energy spectrum in Voigt geometry; note the two Λ-systems with distinct polarization selection rules; d) PL as function of magnetic field in Voigt geometry. (inset: sample and field alignment)

The Voigt geometry data show a fourfold split, with linear polarization, again consistent with donor-bound-exciton emission. From the linesplits, we infer an electron g-factor of 1.2 ($\pm$0.2), which can be compared with the value of 1.36 for a donor-impurity bound electron as obtained by Wolverson *et al.* through spin-flip Raman scattering[36]. The heavy holes in the bound exciton complex are weakly coupled by the magnetic field, leading to an in-plane heavy-hole g-factor ($| \; 3g_{\rm hh}^{\perp} \; |$) of 0.2 ($\pm$0.2). The Voigt data also establish the presence of two doubly-connected Λ-systems, as illustrated in Fig. 3(c), establishing the F:ZnSe system as a candidate for use in several proposed quantum information technology schemes[9–13,37].

In conclusion, we isolated the emission of bound-excitons from a single F:ZnSe complex through mesa-etching and spectral filtering. These methods establish F:ZnSe as a potentially homogeneous and efficient single photon source. The energy splittings from the Faraday and Voigt magneto-photoluminescence data and the presence of TES emission firmly establish the presence of optically controllable electron spins bound to neutral donors in ZnSe. The potential for nuclear purification of the ZnSe host matrix and fabrication ease makes F:ZnSe an attractive candidate for a long-lived, scalable quantum qubit. Future work includes the incorporation of a microcavity[30] to enhance emission and enable efficient quantum networking.

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