Quantum Optical Control of Donor-bound Electron Spins in GaAs

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Summary

Donor-bound electron spins in GaAs are an interesting material system for experimental studies that investigate how quantum information can be stored on the quantum state of a collective spin excitation in an ensemble of spins. Such electron spin ensembles combine reasonably long spin coherence times with well-defined optical transitions, and can therefore be used as a medium for quantum optical studies and work that explores optical quantum-memory functions.

In this thesis we show that this idea is feasible, with experimental work that addresses the donor-bound electron spins with quantum optical techniques. These results provide a basis for future experiments that aim at the preparation and study of nonlocal quantum entanglement between collective spin excitations in two different ensembles, which can be achieved with quantum optical techniques.

In the reported experiments we used the fact that electron spin ensembles can be optically addressed with perpendicular-to-plane propagation of optical control and signal fields. The materials we were using were prepared by standard epitaxial growth techniques for GaAs/$Al_xGa_{1-x}As$ heterostructures. We analyzed that an optimal system is formed in n-GaAs where the level of Si doping is about 10^{14} cm^{-3}. One the one hand, this gives access to a medium with optical density as high as $OD = 1$. This is an important benchmark since the implementation of several quantum memory applications require OD values of 1 and higher. On the other hand, it still assures that each donor-bound electron behaves as an isolated system, without significant interaction with neighboring donor sites.

We can address electron spin degrees of freedom inside ensembles of three-level quantum systems with optical transitions that correspond to the excitation of donor-bound excitons (D^0X systems). These transitions start from the two Zeeman-split spin states of donor-bound electrons (D^0 systems). Selective control over these two transitions is possible with polarization selection rules that naturally occur in this system. In addition, these transitions have very narrow lines and this allows for using spectral selectivity as well.

In order to perform our optical experiments, we have designed and realized a fiber-based confocal microscope that can be used in cryostats with high magnetic field. It is a modular design that can be swapped between use in a helium bath cryostat (4.2 K) and a dilution refrigerator (base temperature well below 20 mK). Faraday rotations in optical materials were circumvented by using a polarization maintaining fiber and by having the light propagating in the sample volume in a direction orthogonal to the applied magnetic field. This also gives access to performing experiments in Voigt geometry, which has several advantages. With experiments on an ensemble of donor-bound electrons in GaAs we confirmed the ability to focus optical control fields with a small spot on any desired point of a sample. We also confirmed that pure linear polarizations can be delivered to the sample, and that this instrument can perform optical experiments at milliKelvin temperatures without excessive heating.

We performed spectroscopy measurements to confirm the optical selection rules and demonstrated the phenomenon Electromagnetically Induced Transparency (EIT). This provides evidence that this medium is suited for the quantum optical techniques that are needed for quantum memory functions and controlled preparation of nonlocal entanglement with ensembles of electron spins. We found that the electron spin dephasing time limits the quality of the EIT. It has the value $T_2^* \approx 2$ ns that results from hyperfine coupling to fluctuating nuclear spins. At the same time, we found that this hyperfine coupling provides a means for controlling the nuclear spin environment via Dynamical Nuclear Polarization (DNP). The EIT spectra form a sensitive probe for detecting how DNP changes the fluctuations and the average of nuclear spin polarization. However, direct optical driving of D^0 transitions yields much weaker DNP effects than that in electron spin resonance experiments with D^0 systems and related optical experiments on quantum dots, and a complete physical picture of DNP effects in our system is not available. Still, initial signatures of controlled DNP effects show that the electron spin-dephasing time can be prolonged. Our experimental approach is suited for exploring this further in conjunction with experiments that aim to implement various applications of EIT

The last chapter of this thesis presents an experiment that studies ultrafast optical preparation of arbitrary coherent dark states of donor-bound spin ensembles, on a timescale that is much shorter than the radiative lifetime of the system. We developed a theoretical model which relies on describing the D^0 systems as 4-level systems (two D^0 levels and two D^0X levels). Within the framework of this model we were able to show that the mechanism of the ultrafast preparation of the coherent dark states relies on a two-photon resonant Raman process. Despite being able to explain most of the results, we had to introduce an ultrafast relaxation time for intra-level relaxation in the D^0X complex, in order to account for the difference in the system's susceptibility to the polarized probe and polarized pump. The existing model and experiments also do not address the question how the coherent dark states evolve when the system initially already had some degree of coherence. More experimental and theoretical investigations need to be done in this direction.

Using this ability to generate the coherent dark states in electron spin ensembles, we were able to make a first systematic study of the spin dynamics in an ensemble of electron spins bound to neutral donors in GaAs, with a time-resolved pump-probe Kerr experiment. Our findings demonstrate that by resonantly exciting the $D^0 - D^0X$ transitions with a single optical pulse, a detectable and well defined electron spin coherence can be generated and its dynamics can be traced in time. We have found that the spin coherence time is strongly dependent on the number of photons in the excitation pulse and in general decreases with increasing power.

The work described in this thesis is a first step towards a variety of quantum optical experiments that can be performed with electron spin ensembles. The observation of EIT allows to pursue an experiment that aims at achieving entanglement between the quantum states of an optical pulse and the state of an electron spin ensemble. Another highly interesting direction aims at extending the spin dephasing time via optical control of the nuclear environment of electron spins. The results in this thesis directly provide all the experimental techniques that are needed for such studies. Extending the spin dephasing time beyond the nuclear-spin-limited $T_2^* = 2$ ns value is of great interest, since it gives access to a much wider scope of experiments that explore quantum optical applications of low-doped *n*-GaAs.