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Introduction

Constructing a quantum computer is one of the challenge of this century. The core component of this computer is the *qbit*, quantum bits. Instead of regular bits, that can take the states (values) $|0\rangle$ and $|1\rangle$, the *qbits*, being quantum devices, can also be in a superposition of states, $\alpha|0\rangle + \beta|1\rangle$. As system able to store the quantum information is therefore searched. The two main criteria for this system is its characteristic time, that must be long enough to do the operation and stock the results, and the ease of preparation in a given state, determining the speed of each operation.

One promising system is the quantum dots (QDs), nanometer sized devices designed to confine carrier in all three dimensions. This confinement leads to a quantization of the carriers energy, akin to the energy level of the electrons in an isolated atom. For this reason, they are dubbed "artificial atom".

Multiple methods exists to form such devices: gate trapping single electrons in-between the electrodes (logical *qubit*), nanometers-sized grains formed by the precipitation of semiconductors in a solution (colloidal dots), thickness variation of a quantum well, strains relaxation of a semiconductor layer... I will focus in this thesis on the later type of QDs, usually grown using Molecular Beam Epitaxy (MBE). They are formed by small island, with a characteristic size of a few nanometers, of a small gap semiconductor inserted in a wide gap semiconductor matrix. Well known example are InAs/GaAs (for the III-V semiconductors), CdSe/ZnSe or CdTe/ZnTe (for the II-VI semiconductors). More specifically, in this thesis, I studied optically active QDs: carriers can be injected in via excitation from a laser, and their relaxation comes with the emission of a photon. Carriers injected in these QDs have a lifetime of a few hundreds of picoseconds [1]. This is too short to stock the informations in any significant amount of time.

The spin coherence time of the carriers spin in these QDs is way longer than the carrier lifetime: the injected electrons can go through several excitation/relaxation while keeping the same spin. It has been shown that, in a single logical *qubit*, the coherence time of the carriers can get as high as 1 μs [2]. Moreover, it has been demonstrated that QDs can be used to control electrically (for the logical *qubit*) or optically (for the optically active dots) the spin of the injected carriers [3, 4].

Optical preparation of the carrier spin state only take a few nanoseconds. All of this makes the spin of carriers trapped in a QD a really promising system for the realization of a *qubit* [5–8]. However, the coherence time of the spin of the carrier sharply drop when an ensemble of QDs is considered, falling to about 10 ns [9–11].

Exiting the world of QDs, two systems can be proposed to get longer spin coherence coherence: Nitrogen-Vacancy (NV) centers [12] or atomic spins directly inserted in the semiconductors [13]. In both those systems, the quantum information can be stored on the spin of the NV center or the magnetic atom. In NV centers, electronic spin coherence in the microseconds range and nuclear spin in the tens of microseconds were found. However, the preparation of the electronic spin of the NV center took hundreds of nanoseconds, which would slow the calculations down. The same kind of coherence and manipulation time can be expected for the atomic spins.

In order to get the best of the two approach, a long coherence with fast preparation, one could imagine to mix those two systems by including magnetic atoms in the QDs. It was shown that, in semiconductors with a low density of magnetic atom, the so called Diluted Magnetic Semiconductors (DMS), there was a strong interaction between the carriers and the magnetic atoms spins. Inserting magnetic atoms in QD, it is possible to use this interaction to control their spins with the injected carriers. In this thesis, the reasoning goes to its limit, inserting a single magnetic atom in a QD, and controlling it optically. Such individual spins are promising for the implementation of emerging quantum information technologies in the solid state [14–16]. Magnetic dopants in conventional semiconductors present many desirable features, such as reproducible quantum properties, stability, and potential scalability for further applications [17]. Thanks to their point-like character, a longer spin coherence time (compared to carriers' spins) can also be expected at low temperature, making them potentially good systems to store quantum information.

The control of the spin state of individual [18–23] or pairs [24, 25] of magnetic atoms has been demonstrated. The spin of a magnetic atom in a QD can be prepared by the injection of spin polarized carriers and its state can be read through the energy and polarization of the photons emitted by the QD [26–28]. The insertion of a magnetic atom in a QD where the strain or the charge states can be controlled also offers degrees of freedom to tune the properties of the localized spin such as its magnetic anisotropy responsible for the spin memory at zero magnetic field [29].

Tab. 1 lists the different magnetic atoms that can be inserted in a semiconductor lattice. Each of those atoms has a unique set of electronic spin, nuclear spin and orbital momentum. Those properties changes the behaviour of the magnetic atom inserted in the semiconductor matrix, and it is therefore interesting to have a large

Table 1: List of different possible transition metals and their key properties in the context of our study.

Inserted atom	V^{2+}	Cr^{2+}	Mn^{2+}	Fe^{2+}	Co^{2+}	Ni^{2+}	Cu^{2+}
d-shell	d^3	d^4	d^5	d^6	d^7	d^8	d^9
Electronic spin	3/2	2	5/2	2	3/2	1	1/2
Nuclear spin	7/2	0	5/2	0	7/2	0	3/2

choice. Mn was the first atom to be successfully inserted and optically probed in CdTe/ZnTe QDs, in 2004 [18]. Since then, several other magnetic atoms have been embedded in II-VI QDs and studied: Co (2014) [30] and Fe (2016) [31].

Mn in II-VI semiconductors has been widely studied in the last decades. In bulk semiconductors, the coherence time was found to reach the milliseconds range for vanishing Mn concentration [32, 33]. Inserted in II-VI QDs, it was demonstrated that a single Mn spin could be optically prepared in a few tens of nanoseconds, depending on the laser power [34]. In the same time, a relaxation time of the Mn spin of a few microseconds was found [26]. The dynamic of a Mn spin was also probed in a positively charged QD, forming a hybrid spin bycoupling with the resident hole [35], and in a strain-free environment [36].

Single Cr atom in a QD is also of particular interest here: its orbital momentum couples to the lattice and make it sensible to the strains. This opens new ways to manipulate the spin state of this magnetic atom without having to use optical excitation. Moreover, it presents no nuclear spin. There is therefore no hyperfine interaction for Cr atom in a CdTe/ZnTe QD. This is expected to lead to longer coherence time for the Cr spin.

In this thesis, I propose to continue the study of the hole-Mn hybrid spin, and to start the study a single Cr atom in a QD. Those two systems are promising for the realization of spin *qubit* coupled to strains. Growth of the Mn-doped QDs was done in Grenoble, in the INAC-CNRS joined team NPSC, by Hervé Boukari. The Cr-doped QDs were grown in Tsukuba, in the team of Pr. Shinji Kuroda, by Hayato Ustumi, Masahiro Sunaga and myself. I studied the dots in Grenoble, with the help of Lucien Besombes.

This thesis is organized as follows:

Chapter I I give in the chapter the theoretical background of this thesis. I begin to discuss the property of a semiconductor crystal. This discussion is then used as a basis to present the physics of the quantum dots and their properties. I leave then the world of QDs to study the interaction between carriers and magnetic atom in a diluted magnetic semiconductor, before applying this to the atoms studied in this thesis: the Mn and the Cr. I also show how the inclusion of these magnetic atom in a crystal affect their spin

energy structure. Finally, I present a short example of application of these theories on the X-Mn system. This theoretical basis will hopefully help the reader to understand the rich physics behind the seemingly simple system of a magnetic atom interacting with an electron and a hole in a QD.

Chapter II The growth of Cr doped QDs was an important part of this thesis. I present here the techniques used to grow the samples studied optically. I begin with a general explanation of the MBE process. I then explain how the Cr-doped QDs were grown and how they are formed using Stranski-Krastanov (SK) relaxation. I also present some tests that were done for the growth of two other kind of sample: charge tunable sample, and strain-free dots formed by the thickness variation of a quantum well. For each kind of sample, I present basic optical characterization.

Chapter III I discuss in this chapter the dynamic of the hole-Mn hybrid spin. I show that optical Λ-level systems appear in the spin structure. Those systems link two hole-Mn ground states to one X^+-Mn excited state via two transitions of opposite polarizations. They were used to study the dynamics of the hole-Mn hybrid spin. A fast h-Mn spin relaxation, in the nanoseconds range, by a coupling with acoustic phonons is evidenced. I also show that two X^+-Mn level are coupled by the in-plane strain anisotropy.

Chapter IV In this chapter, I show that it is possible to include single Cr spin in CdTe/ZnTe QDs and probe its spin optically. The Cr spin structure is deduced from magneto-optical experiments, and shows strong influence of strain. A value of the magnetic anisotropy D_0 between 2 and 3 meV is extracted, two orders of magnitude higher than what is found in Mn-doped QD or in NV centers in diamond. The sign of the hole-Cr exchange interaction is also deduced from these experiments. Finally, I discuss the possibility for a Cr close in the ZnTe barrier close to the QD to influence the QD emission.

Chapter V This chapter explore the dynamics of the Cr spin in a QD. The study begin with photon correlation experiments. In order to get more precise results, pump-probe experiment followed. I begin to present the pump-probe setup, before discussing the results. The success of this experiment shows it the possibility to prepare the Cr spin by optical means. A strong influence of phonons on the Cr spin dynamics is evidenced. A Cr spin relaxation time in the microsecond range is extracted from the experiments. Finally, I also demonstrate the possibility to control the Cr spin by Stark shift.

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