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## **Contrôle optique du spin d'un atome magnétique dans un semiconducteur : spin hybride trou-Manganèse et Chrome**

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But the supervisors and the jury are only a part of the people surrounding you

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# Introduction

Building a quantum computer is one of the challenges of this century. The core component of such a computer is the *qubit*, the quantum bit. Instead of regular bits, which can take the states (values)  $|0\rangle$  and  $|1\rangle$ , the *qubits*, being quantum devices, can also be in a superposition of states,  $\alpha|0\rangle + \beta|1\rangle$ . An important step in the realization of the computer is to find a system to store and control these quantum states, which do not exist yet. The two main criteria for this system are its characteristic time, that must be long enough to do the operation and store the result, and the speed of preparation in a given state, determining the speed of each operation. Moreover, it has to be possible to build gate of one or two *qubits*. The system has also to be scalable, in order to be able to build quantum component with a large number of *qubits*.

Several approaches exist for the fabrication of a *qubit*, such as cold atoms, superconductors... A promising system for their realization is a single quantum dot (QD), nanometer-sized objects designed to confine carriers in all three dimensions. This confinement leads to a quantization of the carrier energy, akin to the energy level of the electron in an isolated atom.

Multiple methods exist to form such devices: gate trapping of single electron between electrodes, nanometer-sized grains formed by the precipitation of semiconductors in a solution (colloidal dots), thickness variation of a quantum well, strain relaxation of a semiconductor layer... I will focus in this thesis on the later type of QDs, usually grown by 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. Well-known examples are InAs/GaAs (for III-V semiconductors), CdSe/ZnSe or CdTe/ZnTe (for II-VI semiconductors). More specifically, in this thesis, I studied optically active QDs: carriers can be injected in the QD by a laser excitation, and their relaxation occurs with the emission of a photon.

The spin of the carriers injected in a QD is a good candidate for the realization of a two level quantum system. For a single gate *qubit*, coherence time of the electrons as long as  $1\ \mu\text{s}$  was found [1]. Moreover, it has been demonstrated that QDs can be used to control electrically (for the gate *qubit*) or optically (for the

optically active dots) the spin of the injected carriers [2, 3]. Finally, the optical preparation of the carrier spin state takes only a few nanoseconds. All of this makes the spin of carriers trapped in a QD a promising system for the fabrication of *qubits* [4–7]. However, the dephasing time of an ensemble of QDs is a lot shorter than the coherence time of single QD, falling to about 10 ns [8–10]. This is too short to do any significant data storage or processing.

Exiting the world of QDs, several systems were proposed to get longer spin coherence time, such as Nitrogen-Vacancy (NV) centers in diamond [11] or atomic spins directly inserted in the semiconductors [12]. In NV centers, electronic spin coherence time in the milliseconds range was found in ultrapure isotopically purified diamonds [13]. However, the preparation of the electronic spin of the NV center takes hundreds of nanoseconds, which would slow the calculations down [11]. The same kind of coherence and manipulation time can be expected for the atomic spins.

Another approach comes from the Diluted Magnetic Semiconductors (DMS). In these materials, a low density of magnetic atoms is inserted in the semiconductor lattice. The semiconductor keeps its conventional optical and electrical properties and new ones arise from the presence of the magnetic atoms. It was shown that there is a strong exchange interaction between the carriers and the magnetic atom spins. When inserting magnetic atoms in a quantum dot, the carriers are confined with them. Their interaction is enhanced, enabling the control of the magnetic atoms spins via the injected carriers. In this thesis, this reasoning is pushed to its limit, inserting a single magnetic atom in a QD, and controlling its spin optically. Such individual spins are promising for the implementation of emerging quantum information technologies in the solid state [14–16]. They were to present many desirable features for the realization of spin *qubits*, such as reproducible quantum properties, stability, and potential scalability by coupling dots [17]. Thanks to their point-like character, a longer spin coherence time (compared to carriers' spins) can also be expected at low temperature. All of this makes single magnetic dopants in QDs a good candidate 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 [29].

Tab. 1 lists some 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. For a given semiconductor structure, those properties



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

Inserted atom	V <sup>2+</sup>	Cr <sup>2+</sup>	Mn <sup>2+</sup>	Fe <sup>2+</sup>	Co <sup>2+</sup>	Ni <sup>2+</sup>	Cu <sup>2+</sup>
<i>d</i> -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

change the magnetic atom behaviour. Each can be used for different applications, such as the realization of a spin mechanical *qubit* for the elements with an orbital momentum. The first atom to have been inserted in a quantum dot is the Mn, first in II-VI (2004) [18], and then in III-V (2007) [20]. Since then, other magnetic atoms have been embedded in II-VI QDs and studied: Co (2014) [30] and Fe (2016) [31]. They have not been inserted successfully in III-V semiconductors yet.

Mn in II-VI semiconductors has been widely studied in the last decades. In bulk semiconductors, its relaxation 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]. At 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 by coupling with the resident hole [35], and in a strain-free environment [36].

Single Cr atom in a QD is also of particular interest: thanks to its orbital momentum, it should be very sensitive to strains. This opens new ways to manipulate the spin state of this magnetic atom without having to use optical excitation. It also opens the possibility to realize spin mechanical system where the Cr is used as a *qubit* coupled to an oscillator. Cr could be used to probe the movement of the oscillator, cool the oscillator down or create non-classical states of the oscillator. Moreover, the Cr atom in a II-VI matrix presents no nuclear spin. There is therefore no hyperfine interaction for Cr atom in a CdTe/ZnTe QD. This simplifies the Cr spin structure and leads to an expected longer coherence time.

In this thesis, I will present a detailed study of the hole-Mn hybrid spin, and to start the study of 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. The optical study of the magnetic QDs was performed at the Néel Institut in Grenoble, where an optical setup for the study of single quantum dots doped with a single magnetic atom had been developed.

This thesis is organized as follows:

**Chapter I** This chapter focuses on the theoretical background of this thesis. I begin to discuss the properties of a semiconductor crystal. This discussion is then used as a basis to present the physics of the QDs and their properties. Then the interaction between carriers and magnetic atom in a diluted magnetic semiconductor is presented. Particular attention is given to the interaction between the carriers and the two atoms studied in this thesis: the Mn and the Cr. I also show how the inclusion of these magnetic atoms in a crystal affects their spin energy structure. Finally, I present a short example of application of these theories on CdTe/ZnTe QDs doped with single Mn.

**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 kinds of Cr-doped samples: 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 spin states form optical  $\Lambda$ -level systems. Two hole-Mn ground states are connected 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 hole-Mn spin relaxation, in the nanoseconds range, caused by the interplay between acoustic phonons and the hole-Mn exchange interaction is evidenced. I also show that two  $X^+$ -Mn level can be coupled by the in-plane strain anisotropy and study this strain induced coherent dynamics.

**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. It is confirmed that the Cr spin is strongly coupled to strains. A value of the magnetic anisotropy  $D_0$  between 2 and 3 meV is extracted. This is 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 extracted from these experiments and found to be anti-ferromagnetic.

**Chapter V** This chapter explore the dynamics of the Cr spin in a QD. The study begins with photon correlation experiments. In order to get more precise results, resonant optical pumping experiments were performed. The optical

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setup is first presented, before discussing the results. The success of this experiment shows the possibility to prepare the Cr spin by spin pumping. A strong influence of phonons on the Cr spin dynamics is evidenced. A Cr spin relaxation time under excitation in the 10 nanoseconds range is extracted from the experiments. In the dark, the relaxation time of the Cr is found to be in the microsecond range. Finally, I also demonstrate the possibility to tune the energy of the Cr spin by optical Stark effect.



# Contents

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<a href="#">Introduction</a>
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