Communauté
UNIVERSITÉ Grenoble Alpes

INSERER LE LOGO DE
L UNIVERSITE PARTENAIRE

THÈSE

Pour obtenir le grade de

DOCTEUR DE LA COMMUNAUTE UNIVERSITE GRENOBLE ALPES

préparée dans le cadre d'une cotutelle *entre la Communauté Université Grenoble Alpes* et …

Spécialité : **Indiquer la spécialité qui figure sur votre carte d'étudiant**

Arrêté ministériel : le 6 janvier 2005 - 7 août 2006

Présentée par

Alban LAFUENTE-SAMPIETRO

Thèse dirigée par **Lucien BESOMBES** et **Shinji SKURODA**
codirigée par **Hervé BOUKARI**

préparée au sein de **l'Institut Néel** et de **l'Université de Tsukuba**

dans **les Écoles Doctorales …**

Titre de la thèse en français

Thèse soutenue publiquement le **26 janvier 2018**,
devant le jury composé de :

Civilité, Prénom, NOM
Fonction et lieu de la fonction, rôle (Président, Rapporteur, Membre)
Civilité, Prénom, NOM
Fonction et lieu de la fonction, rôle (Président, Rapporteur, Membre)
Civilité, Prénom, NOM
Fonction et lieu de la fonction, rôle (Président, Rapporteur, Membre)
Civilité, Prénom, NOM
Fonction et lieu de la fonction, rôle (Président, Rapporteur, Membre)

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 present in this chapter the system we will study as well as the main theoretical tools one needs to understand it. It presents the main theory we need to describe the semiconductor matrix and the quantum dot. We then propose a model for the interaction between the carrier and the magnetic, and apply it to the Mn and the Cr. Finally, we look at how the semiconductor matrix modify the Mn and Cr spin structure.

Chapter II I discuss in this chapter the growth of the quantum dots. Donec

a nibh ut elit vestibulum tristique. Integer at pede. Cras volutpat varius magna. Phasellus eu wisi. Praesent risus justo, lobortis eget, scelerisque ac, aliquet in, dolor. Proin id leo. Nunc iaculis, mi vitae accumsan commodo, neque sem lacinia nulla, quis vestibulum justo sem in eros. Quisque sed massa. Morbi lectus ipsum, vulputate a, mollis ut, accumsan placerat, tellus. Nullam in wisi. Vivamus eu ligula a nunc accumsan congue. Suspendisse ac libero. Aliquam erat volutpat. Donec augue. Nunc venenatis fringilla nibh. Fusce accumsan pulvinar justo. Nullam semper, dui ut dignissim auctor, orci libero fringilla massa, blandit pulvinar pede tortor id magna. Nunc adipiscing justo sed velit tincidunt fermentum.

Chapter III Mauris tempus eros at nulla. Sed quis dui dignissim mauris pretium tincidunt. Mauris ac purus. Phasellus ac libero. Etiam dapibus iaculis nunc. In lectus wisi, elementum eu, sollicitudin nec, imperdiet quis, dui. Nulla viverra neque ac libero. Mauris urna leo, adipiscing eu, ultrices non, blandit eu, dui. Maecenas dui neque, suscipit sit amet, rutrum a, laoreet in, eros. Ut eu nibh. Fusce nec erat tempus urna fringilla tempus. Curabitur id enim. Sed ante. Cras sodales enim sit amet wisi. Nunc fermentum consequat quam.

Chapter IV Quisque consectetuer. In suscipit mauris a dolor pellentesque consectetuer. Mauris convallis neque non erat. In lacinia. Pellentesque leo eros, sagittis quis, fermentum quis, tincidunt ut, sapien. Maecenas sem. Curabitur eros odio, interdum eu, feugiat eu, porta ac, nisl. Curabitur nunc. Etiam fermentum convallis velit. Pellentesque laoreet lacus. Quisque sed elit. Nam quis tellus. Aliquam tellus arcu, adipiscing non, tincidunt eleifend, adipiscing quis, augue. Vivamus elementum placerat enim. Suspendisse ut tortor. Integer faucibus adipiscing felis. Aenean consectetuer mattis lectus. Morbi malesuada faucibus dolor. Nam lacus. Etiam arcu libero, malesuada vitae, aliquam vitae, blandit tristique, nisl.

Chapter V Sed eleifend, eros sit amet faucibus elementum, urna sapien consectetuer mauris, quis egestas leo justo non risus. Morbi non felis ac libero vulputate fringilla. Mauris libero eros, lacinia non, sodales quis, dapibus porttitor, pede. Class aptent taciti sociosqu ad litora torquent per conubia nostra, per inceptos hymenaeos. Morbi dapibus mauris condimentum nulla. Cum sociis natoque penatibus et magnis dis parturient montes, nascetur ridiculus mus. Etiam sit amet erat. Nulla varius. Etiam tincidunt dui vitae turpis. Donec leo. Morbi vulputate convallis est. Integer aliquet. Pellentesque aliquet sodales urna.

Contents

Bibliography

[1] A. Greilich, M. Schwab, T. Berstermann, T. Auer, R. Oulton, D. R. Yakovlev, M. Bayer, V. Stavarache, D. Reuter, and A. Wieck, "Tailored quantum dots for entangled photon pair creation", Phys. Rev. B **73**, 045323 (2006).

[2] J. R. Petta, A. C. Johnson, J. M. Taylor, E. A. Laird, A. Yacoby, M. D. Lukin, C. M. Marcus, M. P. Hanson, and A. C. Gossard, "Coherent manipulation of coupled electron spins in semiconductor quantum dots", Science **309**, 2180–2184 (2005).

[3] A. Greilich, D. R. Yakovlev, A. Shabaev, A. L. Efros, I. A. Yugova, R. Oulton, V. Stavarache, D. Reuter, A. Wieck, and M. Bayer, "Mode locking of electron spin coherences in singly charged quantum dots", Science **313**, 341–345 (2006).

[4] D. Press, T. D. Ladd, B. Zhang, and Y. Yamamoto, "Complete quantum control of a single quantum dot spin using ultrafast optical pulses", Nature **456** (2008).

[5] D. Loss and D. P. DiVincenzo, "Quantum computation with quantum dots", Phys. Rev. A **57**, 120–126 (1998).

[6] A. Imamoḡlu, D. D. Awschalom, G. Burkard, D. P. DiVincenzo, D. Loss, M. Sherwin, and A. Small, "Quantum information processing using quantum dot spins and cavity qed", Phys. Rev. Lett. **83**, 4204–4207 (1999).

[7] D. D. Awschalom and N. Samarth, "Optical manipulation, transport and storage of spin coherence in semiconductors", in *Semiconductor spintronics and quantum computation*, edited by D. D. Awschalom, D. Loss, and N. Samarth (Springer Berlin Heidelberg, Berlin, Heidelberg, 2002), pp. 147–193.

[8] S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnár, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, "Spintronics: a spin-based electronics vision for the future", Science **294**, 1488–1495 (2001).

[9] M. V. G. Dutt, J. Cheng, B. Li, X. Xu, X. Li, P. R. Berman, D. G. Steel, A. S. Bracker, D. Gammon, S. E. Economou, R.-B. Liu, and L. J. Sham, "Stimulated and spontaneous optical generation of electron spin coherence in charged gaas quantum dots", Phys. Rev. Lett. **94**, 227403 (2005).

[10] A. S. Bracker, E. A. Stinaff, D. Gammon, M. E. Ware, J. G. Tischler, A. Shabaev, A. L. Efros, D. Park, D. Gershoni, V. L. Korenev, and I. A. Merkulov, "Optical pumping of the electronic and nuclear spin of single charge-tunable quantum dots", Phys. Rev. Lett. **94**, 047402 (2005).

[11] P.-F. Braun, X. Marie, L. Lombez, B. Urbaszek, T. Amand, P. Renucci, V. K. Kalevich, K. V. Kavokin, O. Krebs, P. Voisin, and Y. Masumoto, "Direct observation of the electron spin relaxation induced by nuclei in quantum dots", Phys. Rev. Lett. **94**, 116601 (2005).

[12] G. D. Fuchs, G. Burkard, P. V. Klimov, and D. D. Awschalom, "A quantum memory intrinsic to single nitrogen–vacancy centres in diamond", Nature Physics **7** (2011).

[13] M. Pierre, R. Wacquez, X. Jehl, M. Sanquer, M. Vinet, and O. Cueto, "Single-donor ionization energies in a nanoscale CMOS channel", Nature Nanotechnology **5** (2009).

[14] M. Veldhorst, C. H. Yang, J. C. C. Hwang, W. Huang, J. P. Dehollain, J. T. Muhonen, S. Simmons, A. Laucht, F. E. Hudson, K. M. Itoh, A. Morello, and A. S. Dzurak, "A two-qubit logic gate in silicon", Nature **526** (2015).

[15] K. Saeedi, S. Simmons, J. Z. Salvail, P. Dluhy, H. Riemann, N. V. Abrosimov, P. Becker, H.-J. Pohl, J. J. L. Morton, and M. L. W. Thewalt, "Room-temperature quantum bit storage exceeding 39 minutes using ionized donors in silicon-28", Science **342**, 830–833 (2013).

[16] N. Bar-Gill, L. Pham, A. Jarmola, D. Budker, and R. Walsworth, "Solid-state electronic spin coherence time approaching one second", Nature Communications **4** (2013).

[17] P. M. Koenraad and M. E. Flatte, "Single dopants in semiconductors", Nature Materials **10** (2011).

[18] L. Besombes, Y. Léger, L. Maingault, D. Ferrand, H. Mariette, and J. Cibert, "Probing the spin state of a single magnetic ion in an individual quantum dot", Phys. Rev. Lett. **93**, 207403 (2004).

[19] C. Le Gall, A. Brunetti, H. Boukari, and L. Besombes, "Optical stark effect and dressed exciton states in a Mn-doped CdTe quantum dot", Phys. Rev. Lett. **107**, 057401 (2011).

[20] A. Kudelski, A. Lemaître, A. Miard, P. Voisin, T. C. M. Graham, R. J. Warburton, and O. Krebs, "Optically probing the fine structure of a single mn atom in an inas quantum dot", Phys. Rev. Lett. **99**, 247209 (2007).

[21] O. Krebs, E. Benjamin, and A. Lemaître, "Magnetic anisotropy of singly mn-doped InAs/GaAs quantum dots", Phys. Rev. B **80**, 165315 (2009).

[22]E. Baudin, E. Benjamin, A. Lemaître, and O. Krebs, "Optical pumping and a nondestructive readout of a single magnetic impurity spin in an InAs/GaAs quantum dot", Phys. Rev. Lett. **107**, 197402 (2011).

[23]J. Kobak, T. Smolenski, M. Goryca, M. Papaj, K. Gietka, A. Bogucki, M. Koperski, J.-G. Rousset, J. Suffczynski, E. Janik, M. Nawrocki, A. Golnik, P. Kossacki, and W. Pacuski, "Designing quantum dots for solotronics", Nature Communications **5**, 3191 (2014).

[24]L. Besombes, C. L. Cao, S. Jamet, H. Boukari, and J. Fernández-Rossier, "Optical control of the spin state of two Mn atoms in a quantum dot", Phys. Rev. B **86**, 165306 (2012).

[25]O. Krebs and A. Lemaître, "Optically induced coupling of two magnetic dopant spins by a photoexcited hole in a mn-doped InAs/GaAs quantum dot", Phys. Rev. Lett. **111**, 187401 (2013).

[26]L. Besombes, H. Boukari, C. L. Gall, B. Brunetti, C. Cao, S. Jamet, and B. Varghese, "Optical control of the spin of a magnetic atom in a semiconductor quantum dot", Nanophotonics **4**, 75 (2015).

[27]D. E. Reiter, V. M. Axt, and T. Kuhn, "Optical signals of spin switching using the optical stark effect in a mn-doped quantum dot", Phys. Rev. B **87**, 115430 (2013).

[28]M. Goryca, T. Kazimierczuk, M. Nawrocki, A. Golnik, J. A. Gaj, P. Kossacki, P. Wojnar, and G. Karczewski, "Optical manipulation of a single Mn spin in a CdTe-based quantum dot", Phys. Rev. Lett. **103**, 087401 (2009).

[29]J. C. Oberg, M. Reyes Calvo, F. Delgado, M. Moro-Lagares, D. Serrate, D. Jacob, J. Fernandez-Rossier, and C. F. Hirjibehedin, "Control of single-spin magnetic anisotropy by exchange coupling", Nature Nanotechnology **9** (2013).

[30]J. Kobak, T. Smoleński, M. Goryca, M. Papaj, K. Gietka, A. Bogucki, M. Koperski, J.-G. Rousset, J. Suffczyński, E. Janik, M. Nawrocki, A. Golnik, P. Kossacki, and W. Pacuski, "Designing quantum dots for solotronics", Nat. Comm. **5**, 3191 (2014).

[31]T. Smoleński, T. Kazimierczuk, J. Kobak, M. Goryca, A. Golnik, P. Kossacki, and W. Pacuski, "Magnetic ground state of an individual Fe^{2+} ion in strained semiconductor nanostructure", Nat. Comm. **7**, 10484 (2016).

[32]D. Scalbert, J. Cernogora, and C. A. L. Guillaume, "Spin-lattice relaxation in paramagnetic CdMnTe", Solid State Communications **66**, 571–574 (1988).

[33]T. Dietl, P. Peyla, W. Grieshaber, and Y. M. d'Aubigné, "Dynamics of spin organization in diluted magnetic semiconductors", Phys. Rev. Lett. **74**, 474–477 (1995).

[34]C. Le Gall, "Dynamics and Optical contrôl of a single spin in a Quantum Dot", Theses (Université de Grenoble, Nov. 2011).

[35]B. Varghese, H. Boukari, and L. Besombes, "Dynamics of a Mn spin coupled to a single hole confined in a quantum dot", Phys. Rev. B **90**, 115307 (2014).

[36]L. Besombes and H. Boukari, "Resonant optical pumping of a Mn spin in a strain-free quantum dot", Phys. Rev. B **89**, 085315 (2014).