Recent optical pumping experiments in semiconductor quantum dots

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Abstract. In this series of lectures, I will treat two different aspects oflight interaction with semiconductor nano-crystals.

I. Rabi oscillations and rapid adiabatic passage in a single quantum dot

After a brief survey of the state of the art techniques to perform micro-spectroscopy of single nano-crystals, I will describe the confined electronic states and the optical selections rules in III-V quantum dots (QD's), showing in particular how to excite a given excited state of the quantum dot from its ground state, and how to detect the resulting population under resonant excitation using photocurrent or a spectator state in luminescence spectroscopy [1,2].

I will then describe the regime of coherent interaction of light with a single quantum dot, which leads to the notion of state dressed by light. This regime can be attained under pulsed resonant excitation, when the light pulse duration is typically shorter than the dephasing-time between the ground and excited state. In a first simple approach, the QD is here described as the two level system constituted by the ground and excited states coupled by light, i.e. the "atom like" description. The possibility to prepare a QD in a given excited state may have strong implications in the improvement of the efficiency of QD-based single photon sources, or polarisation entangled photon pair sources.

I will demonstrate that the achieved QD state preparation at a given excitation power is in fact strongly determined by the light pulse characteristics, as shown in figure 1:
— when the light pulse is Fourier-transform limited, Rabi oscillations appear between the ground and excited state, which are detected as a function of the "pulse-area" Θ, proportional to the integral of the envelope function of the electri cal field of the light pulse [1,3,4].
—when the light pulse is "chirped", which means that the instantaneous light frequency increases (decreases) with time within the pulse, the regime of rapid adiabatic passage can be reached. The conditions to achieve and observe the rapid adiabatic passage will be discussed. It will be shown that, in contrast to Rabi oscillation technique, this regime allows the robust preparation of a given excitation state, here the exciton state [3,4]. A simple model will be presented, based on

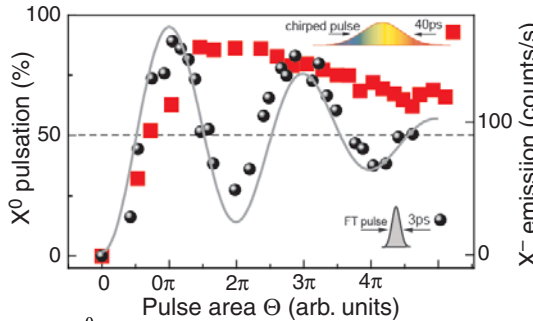

Fig. 1. The exciton X^0 population as a function of the laser pulse area: $\Theta \propto \sqrt{\text{Laser Intensity}}$. The X^0 population is probed by monitoring the negatively charged exciton X^- emission, which is used here as a spectator state. Rabi oscillations (•) of the X^0 population under excitation with Fourier transform limited pulse are observed, in contrast to robust adiabatic passage (square) following excitation with negatively chirped laser pulse stretched to a duration of 40 ps. The decay of the achieved X^0 population (or its oscillation amplitude) is attributed to the coupling by acoustic phonons between dressed exciton states [3,5].

optical Bloch-type equations, which allow a description up to $\Theta \sim 2\pi$.

Finally, I will discuss the limits of the "atom-like" description of the quantum dot, in particular, how the acoustic phonons partly destroy the two level system coherences, both in the Rabi oscillations [5] and in rapid adiabatic passage regimes. The implications for experiments aiming at the coherent control of the dot excitation by light will be evoked [6,7].

II. The electron-nuclear spin system through time or spectral domain spectroscopy

Since the seminal work of G. Lampel in silicon 1], the optical pumping of the nuclear spin system has been the object of extended studies in III-V materials, in particular through optical pumping of electron spin in donor localized states. These studies have been recently renewed thanks to the improvement of growth techniques allowing the elaboration of quantum wells and more recently quantum dots (namely quantum well interface, Stransky-Krastanov, droplet epitaxy QDs) in different semiconductor systems providing new type of tailored confined states, and the possibility to perform spectroscopy either in time domain (mostly on QD ensemble) or in spectral domain where individual nano-crystals can be addressed. These investigations are of potential interest for future highly integrated information storage devices, and more generally for spintronics applications.

I will review and illustrate from experimental examples the different type

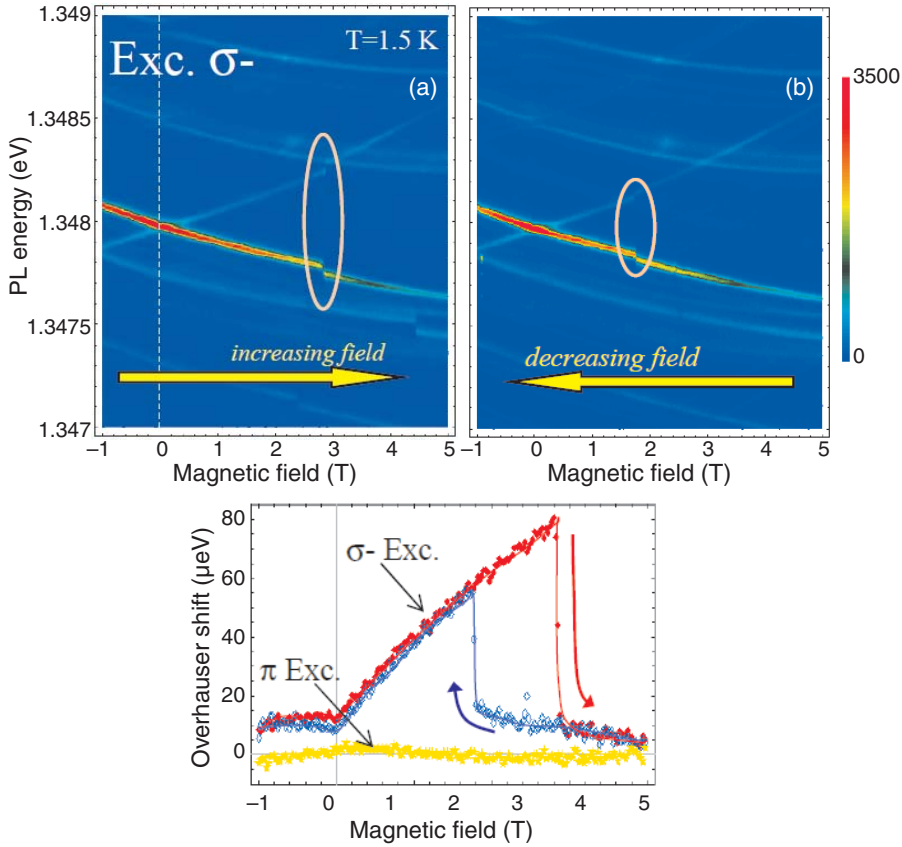

Fig. 2. Bistability of the nuclear field as a function of the longitudinal magnetic field under circularly polarized σ^- cw-excitation of a single dot. (a, b) contour plot of the X^+ charged exciton emission : (a) the nuclear field builds up anti-parallel to the applied field until a critical field ($B_{c2} \sim 3\,\mathrm{T}$) where it abruptly drops down to a small value; (b) decreasing the applied field, the nuclear field rises again abruptly at a critical field ($B_{c2} \sim 1.8\,\mathrm{T}$, $B_{c2} < B_{c1}$). (c) Overhauser shift dependence on the applied magnetic field, showing again the hysteretic loop of the nuclear field [11].

of interactions involving the nuclei [2,3]: spin interactions with external magnetic field, leading to Nuclear Zeeman effect and nuclear magnetic resonance in nanocrystals [4,5]; hyperfine Fermi contact interaction with conduction electron spins, which mediates the optical pumping of nuclear spin by light and are at the origin

of Overhauser and Knight fields, and hyperfine dipolar interaction with valence electron spins. The indirect exchange and dipolar interaction between nuclei will be also mentioned. Finally the quadrupolar interaction, which results from the electrostatic coupling between nuclear spins and the electric field gradients induced by the electrons in the solid, will be described. The latter interaction plays an important part in Stransky-Krastanov QDs, in which strong residual strains remains, a native consequence of the growth technique.

According to the experimental conditions, different type of effects can arise: when the nuclear field is not polarized, the carrier spins can be depolarized by its fluctuations, as evidenced in time resolved photoluminescence of charged exciton X^+ [6] or circular dichroism induced by the resident hole in pump-probe experiments [7,8] performed on QD ensembles. However, in pump-probe Faraday rotation (FR) experiments performed on such ensembles under transverse magnetic field, spin precession of resident electron spin can be modified by local polarization of nuclei, leading to the nuclei-induced frequency focusing of electron spin coherence, which results in long lived electron spin coherences [9]. Under cw-excitation on individual QDs, at high enough pumping rate, it is possible to polarize the nuclei quite efficiently, as revealed by deviations from electron Zeeman effect induced by the nuclear Overhauser field. Different pumping schemes of nuclear spin can arise: the departure from thermal equilibrium of electron spin populations, obtained by non resonant optical pumping of electron spins with circularly polarized light, results in efficient dynamic nuclear polarization (DNP) in Faraday configuration, an effect mediated by the fluctuating hyperfine interaction [10,11], and which can be observed even down to zero external longitudinal magnetic fields in QDs [12]. When the external and the nuclear field are opposite, bistability of the nuclear field can occur (see fig. 2) [10,11,13]. Under transverse magnetic field in Voigt configuration, anomalous Hanle effect is observed, where the nuclear field polarization develops anti-parallel to the applied magnetic field, most likely due to quadrupolar effect in strained QDs, [14,15]. Very efficient DNP mechanism is also obtained under continuous wave resonant optical pumping experiments on single dots by using optically forbidden transitions assisted by hyperfine interaction, termed as the Solid effect [16]. Here, the DNP results directly from the pumping scheme of the nuclei, based on the imbalance between the efficiency of the cycles increasing or decreasing the DNP. Finally, the spectral position of a given optical transition (X^0, $X\pm$) can be strongly modified, typically up to ten times its line width, by DNP under strong magnetic field and resonant excitation by a cw-narrow laser, leading to dragging effect when increasing or decreasing slowly the photon energy [17,18].

References

Part I.

[1] A. Zrenner *et al*., *Nature* **418**, 612 (2002).
[2] Kloeffel *et al*., *Phys. Rev Lett.* **106**, 046802 (2011).
[3] C.-M. Simon *et al*., *Phys. Rev Lett.* **106**, 166801 (2011).
[4] P. R. Eastham *et al*., *Phys. Rev. B* **79**, 165303 (2009); Wu *et al*.., *Phys. Rev Lett.* **106**, 067401 (2011).
[5] A. J. Ramsay *et al*., *Phys. Rev Lett.* **104**, 017402 (2010) ; ibid. **105**, 177402 (2010).
[6] Stufler *et al*., *Phys. Rev. B* **73**, 125304 (2006).
[7] A. J. Ramsay, *Semicond. Sci. Technol.* **25**, 103001 (2010).

Part II.

[1] G. Lampel, *Phys. Rev. Lett.* **20**, 491, (1968).
[2] A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, New York, 1961).
[3] M. I. Dyakonov, *Spin Physics in Semiconductors*, Springer series in solid state science **157**, Springer
[4] D. Gammon, *et al.* , *Science* **277**, 85 (1997).
[5] M. N. Makhonin, *Nature Materials* **10**, 844 (2011).
[6] P. F. Braun *et al*., *Phys. Rev. Lett.* **94**, 116601 (2005).
[7] Eble *et al*., *Phys. Rev. Lett.* **102**, 146601 (2009).
[8] Desfonds *et al*., *Appl. Phys. Lett.* **96**, 172108 (2010).
[9] A. Greilich *et al*., *Science* **317**, 1896 (2007).
[10] P.-F. Braun et al., *Phys. Rev. B* **74** (2006); Urbaszek *et al*.., *Phys. Rev. B* **76**, 201301(R) (2007).
[11] Eble, B. *et al*., *Phys. Rev. B* **74**, 081306(R) (2006); O. Krebs *et al*.., *C.R. Physique***9**, 874 (2008).
[12] Lai *et al*., , *Phys. Rev. Lett.* **96**, 167403 (2006).
[13] Tartakovskii *et al. Phys. Rev. Lett.* **98**, 026806 (2007).
[14] R. Dzhoiev *et al*., *Phys. Rev. Lett.* **99**, 037401 (2007).
[15] O. Krebs *et al*., *Phys. Rev. Lett.* **104**, 056603 (2010).
[16] E. A. Chekhovitch *et al*., *Phys. Rev. Lett.* **104**, 066804 (2010).
[17] C. Latta *et al*., *Nature Physics* **5**, 758 (2009) ; A. Högele *et al*., arXiv :1110.5524v1 (2011).
[18] Yang *et al*., arXiv:1012.0060v2 (2011).