Decoherence and fluctuation dynamics of the quantum dot nuclear spin bath probed by nuclear magnetic resonance

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Decoherence and fluctuation dynamics of the quantum dot nuclear spin bath probed by nuclear magnetic resonance

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Abstract. Dynamics of nuclear spin decoherence and nuclear spin flip-flops in self-assembled InGaAs/GaAs quantum dots are studied experimentally using optically detected nuclear magnetic resonance (NMR). Nuclear spin-echo decay times are found to be in the range 1-4 ms. This is a factor of ~3 longer than in strain-free GaAs/AlGaAs structures and is shown to result from strain-induced quadrupolar effects that suppress nuclear spin flip-flops. The correlation times of the flip-flops are examined using a novel frequency-comb NMR technique and are found to exceed 1 s, a factor of ~1000 longer than in strain-free structures. These findings complement recent studies of electron spin coherence and reveal the paradoxical dual role of the quadrupolar effects in self-assembled quantum dots: large increase of the nuclear spin bath coherence and at the same time significant reduction of the electron spin-qubit coherence. Approaches to increasing electron spin coherence are discussed. In particular the nanohole filled GaAs/AlGaAs quantum dots are an attractive option: while their optical quality matches the self-assembled dots the quadrupolar effects measured in NMR spectra are a factor of 1000 smaller.

1. Introduction

Quantum dots in III-V semiconductors are actively studied for applications in quantum information technologies. The advantages of III-V structures include excellent optical properties and advanced fabrication techniques available for these materials. However, all group III and V atoms have non-zero nuclear magnet moments. Interaction with nuclear spins is a major source of decoherence and information loss in electron and hole spin quantum dot qubits [1]. Nuclear quadrupolar effects and dipolar interactions between nuclei complicate the problem significantly, leading to complex dynamics of the electron-nuclear spin system which lacks complete understanding. Experimental probing of the nuclear spin dynamics is key to building a comprehensive picture of the electron-nuclear spin physics in quantum dots. In what follows measurements of the different timescales characterizing the nuclear spin bath are presented and discussed.

2. Nuclear spin dynamics in semiconductor quantum dots

2.1. Experimental procedure

We use optically detected nuclear magnetic resonance (ODNMR) to study a variety of quantum dot systems: self-assembled InGaAs/GaAs and InP/GaInP dots [2], monolayer fluctuation GaAs/AlGaAs dots [3] as well as GaAs/AlGaAs quantum dots grown by nanohole etching and in-filling [4]. All

experiments are performed at T=4.2 K in magnetic fields up to B_z=10 T along the growth direction of the sample. Key to ODNMR measurement is the hyperfine interaction between electron and nuclear spins [1], which plays a dual role. Firstly, when quantum dot is excited with circularly polarized light, spin polarized electrons are generated repeatedly and hyperpolarize the nuclei in turn. Secondly, polarized nuclear spins produce an effective (Overhauser) magnetic field which manifests itself in the hyperfine shift E_{hf} of the electron spin levels and can be detected in the luminescence spectra – this allows ODNMR sensitivity down to individual quantum dot level.

2.2. Longitudinal nuclear spin relaxation ($T_{1,nucl}$)

Longitudinal relaxation time $T_{1,nucl}$ of the nuclear spin magnetization in the dark (without optical excitation) can be probed with optical "pump-probe" spectroscopy. Extremely long relaxation times exceeding 1000 s have been observed in self-assembled quantum dots [5, 6] and attributed to suppression of the dipolar nuclear flip-flops arising from large inhomogeneous strain induced quadrupolar effects. In strain-free GaAs/AlGaAs quantum dots the relaxation times are shorter (~60 s) but are still longer than in bulk semiconductors due to spin diffusion suppression [7]. Such $T_{1,nucl}$ times are much longer than any electron spin timescales in quantum dots implying that longitudinal nuclear spin relaxation does not limit electron spin coherence.

2.3. Transverse nuclear spin relaxation ($T_{2,nucl}$)

Unlike the longitudinal component, nuclear magnetization perpendicular to the external field is more prone to relaxation, since such process does not require energy exchange with environment. The transverse relaxation time of the nuclear magnetization $T_{2,nucl}$ is measured using Hahn echo pulse sequence [8, 9] shown in the inset of Fig. 1. The role of the π-pulse is to refocus the dephasing caused by inhomogeneous NMR spectral broadening, so that $T_{2,nucl}$ is a measure of the nuclear dipolar interactions only. The curves in Fig. 1 show the amplitude of the spin echo as a function of the evolution time 2τ measured for ^{69}Ga and ^{115}In nuclei of a single self-assembled InGaAs quantum dot. The echo decay times $T_{2,nucl}$ derived from fitting are noticeably larger than in strain-free structures (e.g. for ^{69}Ga nuclei we find $T_{2,nucl}$ =2.1 ms compared to $T_{2,nucl}$ =0.36 ms in strain-free dots [8]). Such increase in $T_{2,nucl}$ in self-assembled dots is due to the large inhomogeneous quadrupolar effects which shift the nuclear spin levels. Such shifts result in suppression of the dipolar flip-flops (flip-flop is a process where two nuclei exchange nuclear magnetization parallel to the magnetic field) as they become energetically forbidden.

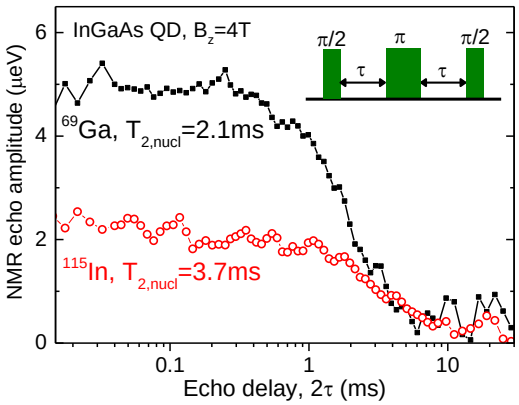

Figure 1. Nuclear spin echo decay in a single self-assembled quantum dot. Inset shows the Hahn echo pulse sequence.

On the other hand, upon closer examination, one finds that even complete freezing of the nuclear spin flip-flops does not lead to infinite nuclear spin coherence. Instead there is a maximum value of $T_{2,nucl}$ that can be calculated using the method of moments. For example, for ^{69}Ga the maximum $T_{2,nucl}$ is 3.0ms [9], thus experimental observation of a close to the limit $T_{2,nucl}$ =2.1 ms suggests that dipolar flip-flops are strongly suppressed in self-assembled dots. However, the degree of such suppression is difficult to gauge – this is an intrinsic feature of the spin echo and pulsed NMR in general. The upper limit on $T_{2,nucl}$ can be viewed as arising from parasitic spin dynamics induced by the "hard" pulses inducing the spin echo (an effect known as "instantaneous diffusion"). Probing equilibrium nuclear spin dynamics, which are of most interest, requires alternative NMR techniques discussed below.

2.4. Using frequency comb NMR to measure homogeneous NMR linewidth ($\Delta\nu_{hom}$)

Here we consider an alternative to pulsed NMR where low power radio frequency (rf) signals inducing non-coherent dynamics (without Rabi oscillations) are used. In the case of self-assembled quantum dots the inhomogeneous NMR spectrum of each isotope with linewidth $\Delta\nu_{inh}$ consists of a large number of individual NMR transitions with homogeneous linewidth $\Delta\nu_{hom} << \Delta\nu_{inh}$ (sketched with thick lines in Fig. 2(a)). Radio frequency signals with frequency comb profiles are used (thin lines in

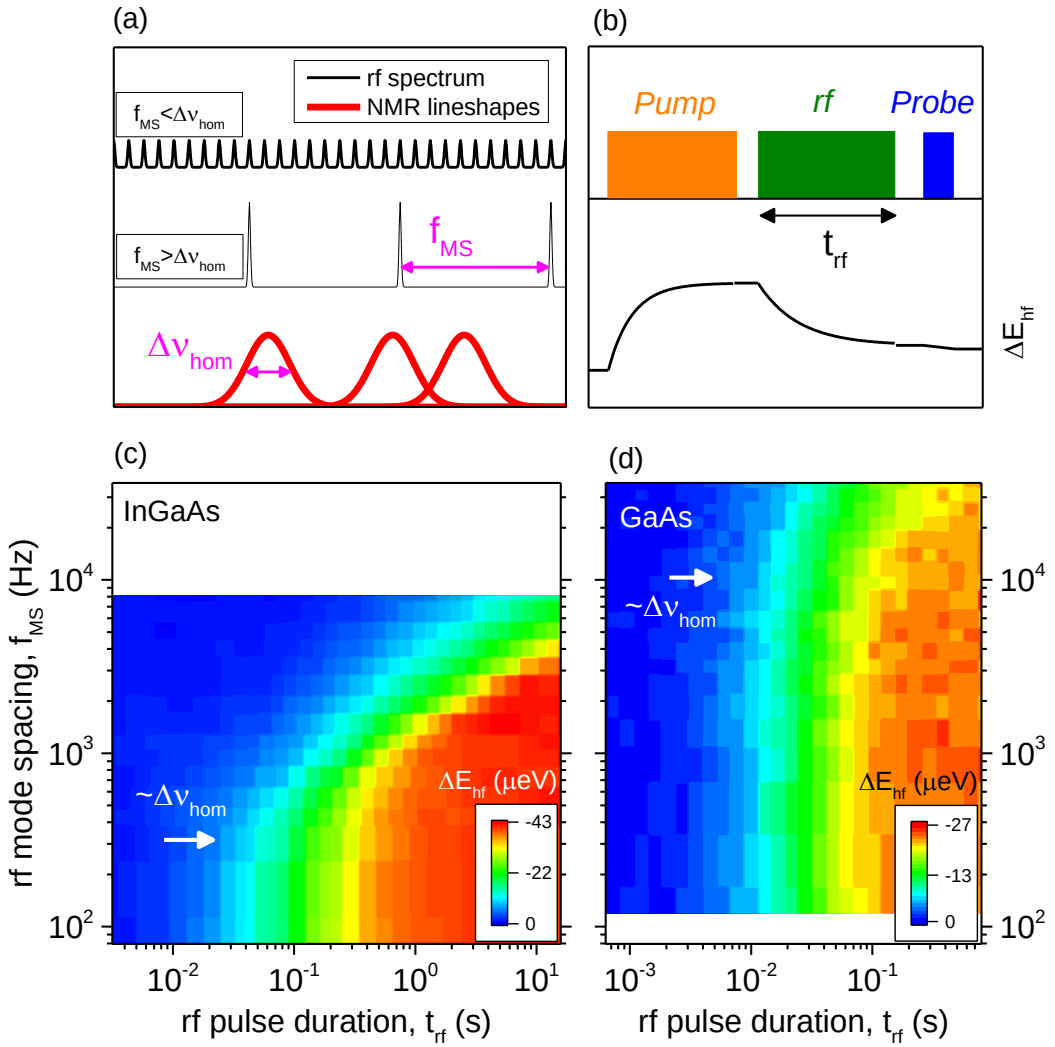

Figure 2. (a) Schematic spectra of NMR transitions (thick lines) and frequency combs (thin lines). (b) Time diagram of the measurement with a sketch of the corresponding change in the hyperfine shift ΔE_{hf}. (c, d) Experimental depolarization of ^{75}As spins in an InGaAs/GaAs (c) and GaAs/AlGaAs (d) quantum dot.

Fig. 2(a)). Depending on the mode spacing of the comb f_{MS} two opposite cases arise: (i) For $f_{MS} << \Delta\nu_{hom}$ all NMR transitions are uniformly excited and fast depolarization is expected, (ii) for $f_{MS} >> \Delta\nu_{hom}$ some of the NMR transitions are out of resonance with the comb, so that a slowdown in depolarization is expected. Thus non-coherent nuclear spin depolarization is sensitive to $\Delta\nu_{hom}$.

In experiment (Fig. 2(b)) the nuclei are first polarized optically. This is followed by depolarization with a frequency comb rf pulse in the dark (duration t_{rf}). Finally nuclear magnetization is probed by measuring the change in the hyperfine shift ΔE_{hf} in the photoluminescence spectrum. The dependence $\Delta E_{hf}(t_{rf},f_{MS})$ is shown in Fig. 2(c) for the ^{75}As nuclei of an InGaAs quantum dot. As expected, at small

f_{MS} depolarization is fast. But above a certain threshold value of f_{MS} a pronounced slowdown is observed – this threshold in f_{MS} gives a few hundred Hz estimate of $\Delta\nu_{hom}$. A more accurate value of the homogeneous linewidth $\Delta\nu_{hom}\approx 210$ Hz is obtained from the rate equation modelling [10]. The corresponding $T_{2,nucl}\approx 1/(\pi\Delta\nu_{hom})\approx 1.5$ ms is in reasonable agreement with the spin echo measurement [9]. A significantly different result is observed in Fig. 2(d) for a nearly strain-free monolayer fluctuation GaAs/AlGaAs quantum dot: the slowdown in nuclear depolarization is observed only for the comb mode spacing f_{MS} >10kHz, with fitting revealing much larger $\Delta\nu_{hom}\approx 3200$ Hz and hence smaller $T_{2,nucl}\approx 0.1$ s. Such homogeneous broadening is due to energetically allowed nuclear dipolar flip-flops and possibly due to some additional effect of charge fluctuations.

It is worth noting that even with completely frozen dipolar flip-flops, the homogeneous linewidth $\Delta\nu_{hom}\approx 210$ Hz obtained for ^{75}As in an InGaAs quantum dot is smaller than the homogeneous broadening expected for all possible states of the nuclear spin environment surrounding each ^{75}As spin. This important feature of the frequency comb technique arises from the fact that the nuclear spin environment is sampled over a finite timescale, comparable to the exponential depolarization time achieved at $f_{MS} << \Delta\nu_{hom}$. Thus for sufficiently fast frequency comb rf depolarization the environment of each depolarized spin is effectively static ("frozen") and the dipolar field produced by such environment is eliminated as any other source of inhomogeneous broadening. As described below this property of the frequency comb NMR method opens a way for direct probing of the equilibrium flip-flop dynamics in self-assembled quantum dots.

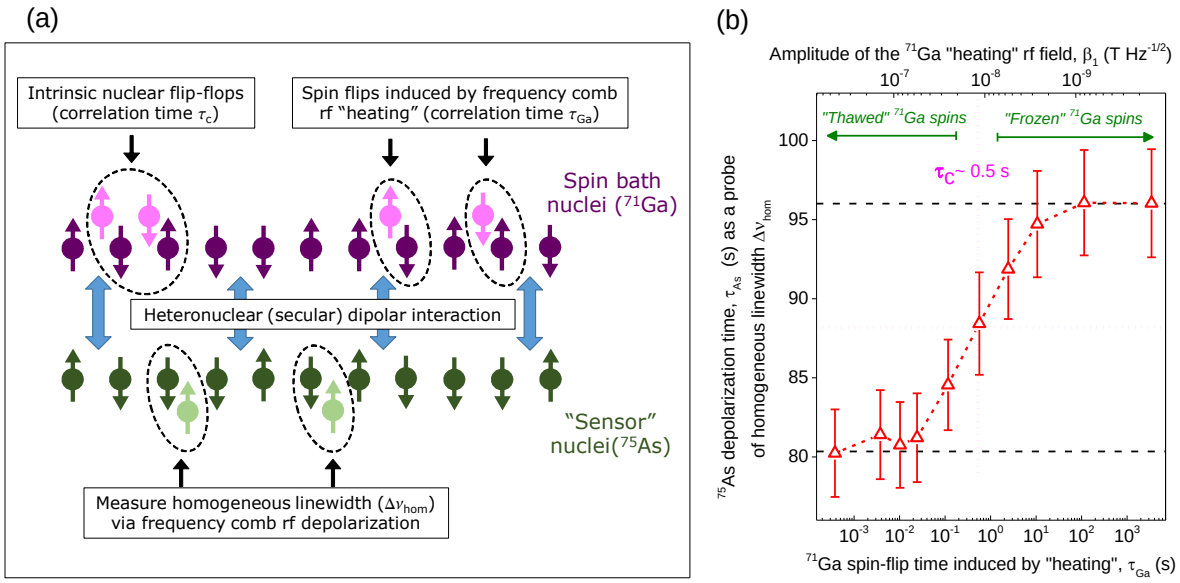

Figure 3. (a) Measurement protocol to determine correlation time τ_c of the nuclear dipolar flip-flops. (b) Experimental dependence of the ^{75}As rf-induced depolarization time on the amplitude of the ^{71}Ga rf heating amplitude. The half-range point (dotted lines) corresponds to the situation where natural flip-flops and rf heating induced spin flips of ^{71}Ga have the same effect on ^{75}As, revealing the correlation time τ_c of the ^{71}Ga flip-flops.

2.5. Nuclear spin flip-flop correlation times ($\tau_{c,nucl}$)

The strategy for probing the correlation time $\tau_{c,nucl}$ of the nuclear spin exchange flip-flops is shown in Fig 3(a). The ensemble of ^{71}Ga spins is studied, while ^{75}As nuclei are used as a "sensor". Heteronuclear (^{71}Ga-^{75}As) dipolar interaction is secular: the ^{71}Ga-^{75}As flip-flops are energetically forbidden but random configurations of the ^{71}Ga spins cause homogeneous broadening of the ^{75}As nuclear spins. The ^{71}Ga spins undergo random flip-flops with unknown correlation time $\tau_{c,nucl}$. This fluctuation correlation time can be shortened by an additional "heating" with a frequency comb rf (resonant with ^{71}Ga) which produces spin flips with a known correlation time τ_{Ga}. A pronounced speed

up of the ^{71}Ga fluctuations occurs for $\tau_{Ga} \approx \tau_{c,nucl}$ and is detected via increased $\Delta\nu_{hom}$ of the ^{75}As spins. This allows for $\tau_{c,nucl}$ to be derived. Measuring the entire $\Delta E_{hf}(t_{rf}, f_{MS})$ dependence of ^{75}As is time consuming, however, in order to characterize the changes in $\Delta\nu_{hom}$ it is sufficient to measure the characteristic depolarization time τ_{As} at a fixed $f_{MS} \approx \Delta\nu_{hom}$. The resulting $\tau_{As}(\tau_{Ga})$ dependence is shown in Fig. 3(b): the transition point $\tau_{Ga} \approx \tau_{c,nucl}$ is approximately at the middle of the τ_{As} variation range (dotted lines) providing an estimate $\tau_{c,nucl} \approx 0.5$ s, with a more accurate $\tau_{c,nucl} \approx 1$ s obtained from model fitting [10].

The flip-flop correlation times $\tau_{c,nucl} \geq 1$ s derived from frequency comb NMR of neutral self-assembled quantum dots are very long and are a sign of extreme freezing of the flip-flops induced by inhomogeneous quadrupolar shifts. Nuclear dipolar flip-flops are responsible for the "spectral diffusion" process [11] which sets an ultimate upper limit on the coherence of the central (electron or hole) spin. It is thus evident that intrinsic nuclear spin dynamics of a bare quantum dot do not limit the electron spin coherence time $T_{2,elec}$ up to sub second regime. A more complex picture is expected when electron-nuclear interactions are present. This is a subject for a further investigation using frequency comb NMR on electron charged quantum dots.

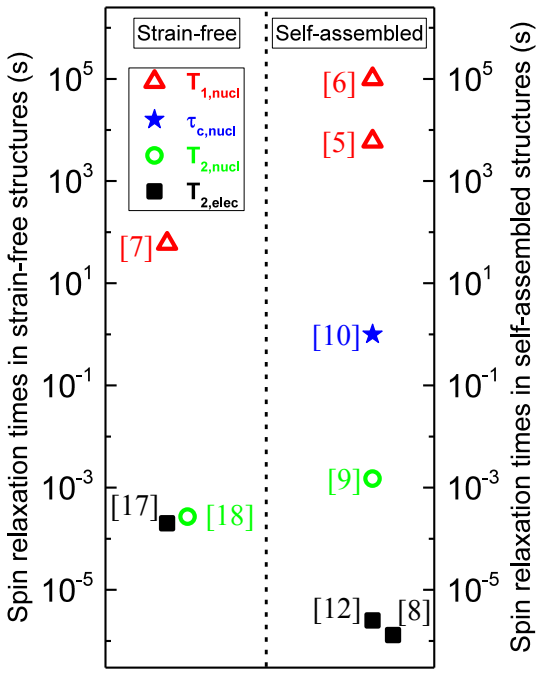

Figure 4. Electron and nuclear spin relaxation times in strain-free GaAs and self-assembled InGaAs quantum dots. Numbers show corresponding references.

3. Discussion and outlook

3.1. Comparison of electron and nuclear spin relaxation timescales

The various experimentally measured timescales characterizing the nuclear spin as well as electron spin dynamics are shown in Fig. 4 summarizing the recent studies for both strain-free lattice matched GaAs/AlGaAs and self-assembled strained InGaAs/GaAs quantum dots.

In strain-free structures electron spin coherence times $T_{2,elec}$ and nuclear spin echo times $T_{2,nucl}$ of few hundred microseconds are reported. Although not measured directly the flip-flop correlation times are expected to be comparable to the coherence times $\tau_{c,nucl} \approx T_{2,nucl}$ in structures with negligible quadrupolar effects. Thus one concludes that in the strain-free GaAs quantum dots the electron spin qubit coherence is limited by the nuclear spin fluctuation dynamics.

A very different and somewhat paradoxical picture is observed for self-assembled dots. Large quadrupolar effects result in a dramatic increase of all nuclear spin relaxation timescales. On the other hand, the electron spin coherence times are found to be much shorter than in strain-free quantum dots. There is a striking six orders of magnitude gap between electron spin coherence time $T_{2,nucl}$ and nuclear spin correlation time $\tau_{c,nucl}$. While initial studies considered extrinsic limitations to electron spin coherence (such as charge fluctuations in quantum dot environment [12]), more recent reports point to intrinsic limitations. The proposed mechanisms include electron mediated nuclear-nuclear interactions [13] as well as broadband nuclear spin noise arising from large inhomogeneous quadrupolar effects [14]. Building a complete picture of the factors limiting the electron spin coherence in self-assembled dots requires further studies: NMR techniques reported here are expected to be a valuable tool in such work. Meanwhile it is important to consider the available approaches to increasing the quantum dot spin qubit coherence as discussed below.

3.2. Approaches to increase quantum dot spin coherence

Recent proposals to improve qubit coherence in self-assembled dots include switching from electrons to valence band holes which have much smaller hyperfine interaction [15], as well as initializing the nuclear spin bath in an eigen state by achieving nuclear spin polarization close to 100% [16].

One more approach is to eliminate quadrupolar effects altogether: This is indeed possible in strain-free quantum dots in 2DEG in GaAs, however, such structures lack the optical access to the spin of the electron. On the other hand, a recently developed growth method based on in situ etching and filling of the nanoholes yields GaAs/AlGaAs quantum dots that combine the excellent optical properties of the self-assembled dots but with very small quadrupolar effects. Recent NMR measurements have shown that quadrupolar shifts in such dots are within ±20 kHz, which is three orders of magnitude smaller than in InGaAs dots [4]. Furthermore, application of external strain cancels the average quadrupolar shifts leaving only the effects of inhomogeneous NMR broadening that does not exceed ±10 kHz. A direct measurement of electron spin coherence in such dots could be very revealing in achieving deeper understanding of the electron-nuclear spin dynamics and the role of the quadrupolar interactions.

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