

NUCLEAR SPIN RELAXATION AND SPIN SQUEEZING UNDER ELECTRIC QUADRUPOLE INTERACTION

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Nuclear Spin Relaxation And Spin Squeezing Under Electric
Quadrupole Interaction

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We certify that we have read this thesis and that in our opinion it is fully adequate,
in scope and in quality, as a thesis for the degree of Master of Science.

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ABSTRACT

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Nuclear spins dynamics recently gained prominence for semiconductor quantum information technologies. At least two ramifications can be mentioned within this context: first, as a decoherence channel for carrier spin qubit stored in a quantum dot, and second as a potential quantum memory with the proviso that the nuclear spin bath can be tamed. To shed light on either of these matters, this thesis presents numerical simulations of spin dynamics of quadrupolar nuclei which constitute a large fraction of group III-V semiconductors. Particular attention is devoted to the electric quadrupole interaction that prevail in these strained semiconductor structures. Within Lindblad master equation formalism, the saturation under an incoherent radio frequency pump, and subsequent relaxation of spin-3/2 nuclei are studied. The quadrupole interaction does not manifest itself via a conspicuous fingerprint in these processes other than causing faster relaxation. However, we identify that its prime role is in spin squeezing. The characteristics of all spins between 1/2 to 9/2 have been thoroughly investigated under one-axis, mixed-axis, and two-axis countertwisting conditions. Our main conclusion is that the presence of quadrupole interaction substantially degrades the average level of squeezing, which further complicates the quantum control of nuclear spin bath fluctuations.

Keywords: Nuclear spin dynamics, spin relaxation, electric quadrupole interaction, spin squeezing.

ÖZET

ELEKTRİKSEL DÖRTKUTUP ETKİLEŞİMİ ALTINDA ÇEKİN SPİN GEVŞEMESİ VE SPİN SIKIŞTIRILMASI

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Çekin spin devinimi, son dönemlerde kuantum bilgi teknolojileri kapsamında önem kazanmaya başlamıştır. Bu bağlamda, en az iki ayrı husustan söz edilebilir: ilki, kuantum noktada saklanan taşıyıcı spin kübitinin eşevresizliğine neden olması, ve ikincisi spin hamamının denetimiyle potensiyel kuantum hafızasının sağlanması. Bu tez, yukarıdaki içeriklerden herhangi birini aydınlatmak için, Grup III-V yarıiletkenlerinin büyük bir kısmını oluşturan dörtkutuplu çekin spin deviniminin nümerik simülasyonunu içermektedir. Bu ilginin bir kısmı, yarıiletken yapılardaki gerilimin ortaya çıkardığı elektriksel dörtkutup etkileşimine yöneliktir. Lindblad egemen denklem çözümlemesi içerisinde spin- $3/2$ çekirdeği için, eşevreli olmayan radyo frekanslı pompa altında doyuma ulaşma ve bunu takip eden gevşeme süreçleri çalışılmıştır. Bu süreçlerde, dört kutup etkileşimi daha hızlı bir gevşeme dışında göze çarpan bir biçimde kendisini göstermemektedir. Tek, karışık, ve çift eksenli karşı burkma koşulları altında, $1/2$ ve $9/2$ arasındaki bütün spin değerleri incelenmiştir. Genel sonucumuz, dörtkutup etkileşiminin varlığının, sıkıştırılmayı büyük ölçüde azalttığı yönündedir ki bu, çekin spin hamamındaki dalgalanmaların kuantum kontrolünü karmaşıktırmaktadır.

Anahtar sözcükler: Çekin spin devinimi, spin gevşemesi, elektriksel dörtkutup etkileşimi, spin sıkıştırılması.

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Chapter 1

Introduction

With the advancement of our mastery on quantum mechanics and its applications, together with the ever increasing computing needs gave rise to the idea of quantum computers toward the turn of the century. As one of the scalable approaches, semiconductor quantum dots (QDs) bring into play the nuclear spins that can be manipulated to store information just by using external magnetic field [1]. Following this proposal, nuclear spins started to be used in quantum communications [2] and quantum networks [3]. Foremost, a quantum computer needs a quantum memory to store a qubit for long enough times, where nuclear spins can be employed [4]. For instance, the quantum state of an electron can be stored in nuclear spins that can be used in quantum memories [5]. As a matter of fact, the single qubit system in QDs lets the time long enough for quantum computation [6, 7]. Therefore, controlling the spin qubits in QDs is vital for taming decoherence [8, 9, 10]; the entanglement in the qubits helps information transfer and changing the electron spins make possible the measurements that cannot be done classically [11].

These few examples show us why controlling nuclear spins and their relaxation are important. One of the milestone studies on spin relaxation is Solomon's work published in 1955 [12]. In this paper, he formulated the relaxation process of two-spin system via a Hamiltonian including the Zeeman and dipole-dipole

interactions. Shortly afterwards, Redfield went on studying relaxation including the quadrupolar interaction in 1957 [13]. After these seminal works, studies on relaxation boomed in parallel with its importance in nuclear magnetic resonance (NMR) [14, 15, 16, 17]. The so-called spin-lattice relaxation is the fundamental concept in NMR experiments that include magnetic and quadrupolar interactions [14]. Also, this type of relaxation helps to understand the processes in QDs under a magnetic field [17]. One of the papers published on NMR experiments [16] focuses on the relaxation process of nuclear spin $3/2$ using Redfield theory. This paper forms a representative example for the type of relaxation measurements that we would like to shed light on.

Another central topic for this thesis is spin squeezing. The pioneering work in this direction belongs to Kitagawa and Ueda [18]. They propose spin squeezing with one-axis twisting, or two-axis countertwisting for canceling out the fluctuations in a certain direction and hence suppressing noise. From this starting point, numerous other papers both experimental and theoretical in different subjects related to spin squeezing have been published [19, 20, 21, 22]. There are a number of reasons spin squeezing is used this frequently through years. Spin squeezing is used to detect many-particle entanglement [23]. Also, spin squeezing helps to increase the precision in measurements [24]. It has other applications on quantum computing, quantum simulation etc.[25, 26, 27, 28] The spin squeezing is used in steady-state entanglement to increase fidelity which is important for quantum memories [26]. Also, it is used in spin bath to combat the decoherence process [28]. There are studies on NMR systems with quadrupolar nuclei using spin squeezing, aiming to make a development in quantum computers [22]; this very recent experiment forms an inspiration for our interest on squeezing that gave rise to the second part of this thesis.

1.1 This Thesis

In this thesis, first, we aim to understand the saturation and subsequent relaxation of a spin $I = 3/2$ system. Our choice of $I = 3/2$ nuclear spins is due to

dominance of these nuclei in III-V semiconductor QDs like Ga and As. Our system is an open four-level cascade system. It is pumped with an incoherent resonant radio frequency field. For relaxation and saturation of the system, Zeeman and quadrupolar interactions are taken into account. The open system is characterized by transitions via magnetic dipolar and electric quadrupolar environmental fluctuations. For this purpose we resort to the Lindblad Master Equation.

In the second part of the thesis, we again use the quadrupole interactions, but this time to drive the spin squeezing mechanism for examining the change in the fluctuations of the quadratures while obeying the Heisenberg limit for their product. Unlike the one-axis and two-axis countertwisting Hamiltonians considered frequently in the literature [19], we explore the mixed-axis twisting which is the native quadrupole Hamiltonian. We compare all these squeezing Hamiltonians to reveal how the squeezing evolves over time in each case.

For the type of spin dynamics required in this work, we benefited from the python library QuTiP developed by P.D. Nation and J.R. Johansson [29, 30], which provides a convenient infrastructure not only for numerics but also for its visualization.

Chapter 2

Elements of Theory

This chapter contains elements of theory to be employed in the remainder of the thesis. We begin with the Lindblad master equation which is predominantly employed in our analysis of relaxation, specifically in four-level systems. Then, we discuss the electric quadrupole interaction which is followed by coherent spin states and measures for spin squeezing. We are rather concise in our treatment, and refer to sources that contain extensive discussions on each of these subjects.

2.1 Lindblad Master Equations

A system S that is interacting with other system(s) having a large degree of freedom, usually abbreviated as environment E , is described by the joint density matrix $\hat{\rho}_{SE}(t)$. If one considers independent initial conditions we write the density operator (matrix) using tensor product as

$$\hat{\rho}_{SE}(t_0) = \hat{\rho}_S(t_0) \otimes \hat{\rho}_E(t_0). \quad (2.1)$$

To get the density matrix for only the system $\hat{\rho}_S(t)$, we trace out over the environment. The result is called the *reduced density matrix* of the system given by

$$\begin{aligned}
\hat{\rho}_S(t) &= \text{Tr}_E \{ \hat{\rho}_{SE}(t) \} , \\
&= \text{Tr}_E \left\{ \hat{U}(t, t_0) \hat{\rho}_{SE}(t_0) \hat{U}^\dagger(t, t_0) \right\} , \\
&= \text{Tr}_E \left\{ \hat{U}(t, t_0) \hat{\rho}_S(t_0) \otimes \hat{\rho}_E(t_0) \hat{U}^\dagger(t, t_0) \right\} ,
\end{aligned} \tag{2.2}$$

where $\hat{U}(t)$ is the time evolution operator and equals to $\exp \left\{ -i \hat{H}_{SE}(t - t_0) / \hbar \right\}$.

However, this form is computationally highly formidable, if not impossible, as it requires full dynamics of both the system *and* the environment. At this point, the Lindblad master equation comes to rescue, by offering a viable approximation for the time evolution of the reduced system density matrix. It is a type of Markovian master equation where memory effects of the environment E are ignored [31]; the form that is usually preferred in computations is expressed as

$$\frac{d}{dt} \hat{\rho}_S(t) = -i \left[\hat{H}'_S, \hat{\rho}_S(t) \right] - \frac{1}{2} \sum_{\mu} \kappa_{\mu} \left\{ \hat{\mathcal{L}}_{\mu}^{\dagger} \hat{\mathcal{L}}_{\mu} \hat{\rho}_S(t) + \hat{\rho}_S \hat{\mathcal{L}}_{\mu}^{\dagger} \hat{\mathcal{L}}_{\mu} - 2 \hat{\mathcal{L}}_{\mu} \hat{\rho}_S(t) \hat{\mathcal{L}}_{\mu}^{\dagger} \right\} , \tag{2.3}$$

where, $\hat{H}'_S$ is the renormalized Hamiltonian of the system; the legacy of interaction with the environment being $\hat{\mathcal{L}}_{\mu}$ carrying the names Lindblad operators, or collapse and sometimes jump operators, with κ_{μ} being the corresponding rate coefficients of these jump events. In the next section, we shall give concrete examples for these Lindblad operators. An extensive discussion of decoherence processes and their mathematical modelling can be found in Schlosshauer's recent book [31].

2.2 Quadrupolar Interaction

Nuclei with spins $I > 1/2$ possess non-spherical charge distributions, hence they have a non-zero electric quadrupole moment, Q [32, 33]. Therefore, they couple to the so-called electric field gradient (EFG) [34], if available at the nuclear site. In a solid-state context, one common cause is the crystal electric fields of polar group III-V semiconductor QDs under inhomogeneous strain [35].

The elements of the EFG tensor is given by the spatial second derivatives of

the (crystal) electric potential V

$$V_{ij} \equiv \frac{\partial^2 V}{\partial x_i \partial x_j}. \quad (2.4)$$

The convention is to label the coordinate axes such that $|V_{xx}| \leq |V_{yy}| \leq |V_{zz}|$, where z is referred to as the the major principal axis of the EFG tensor [32, 33].

There are two important parameters of EFG: eq and η [32, 33]

- eq : This parameter is the major principal value of the EFG and equals to

$$eq = V_{zz},$$

- η : This parameter is the asymmetry of the EFG

$$\eta = \frac{(V_{xx} - V_{yy})}{V_{zz}},$$

together with the condition $\sum_i V_{ii} = 0$ [32, 33], causes η to range between 0 and 1.

If the quadrupolar effect is smaller than the Zeeman interaction, then in perturbation only the first-order quadrupolar Hamiltonian $\hat{H}_Q^{(1)}$ suffices. For larger quadrupolar effect, one should also add the second-order quadrupolar Hamiltonian $\hat{H}_Q^{(2)}$. This will be the basis for the one- and two-quantum quadrupole transitions to be introduced in the next section. The full quadrupole Hamiltonian is given by [32]

$$\hat{H}_Q = \frac{e^2 q Q}{4I(2I-1)} \left[3\hat{I}_z^2 - \hat{I}^2 + \eta \frac{\hat{I}_+^2 + \hat{I}_-^2}{2} \right]. \quad (2.5)$$

Alternatively, if we insert $\hat{I}_\pm \equiv \hat{I}_x \pm i\hat{I}_y$ in the equation above, we get the form

$$\hat{H}_Q = \frac{eQ}{4I(2I-1)} \left[V_{zz}(3\hat{I}_z^2 - \hat{I}^2) + (V_{xx} - V_{yy})(\hat{I}_x^2 - \hat{I}_y^2) \right] \quad (2.6)$$

Here, e is the electron charge, q is the major field gradient and Q is the quadrupole moment, as mentioned above.

For further details, we refer to the historical references [32, 33] that remain to be the most comprehensive treatments. We shall return to the full quadrupole

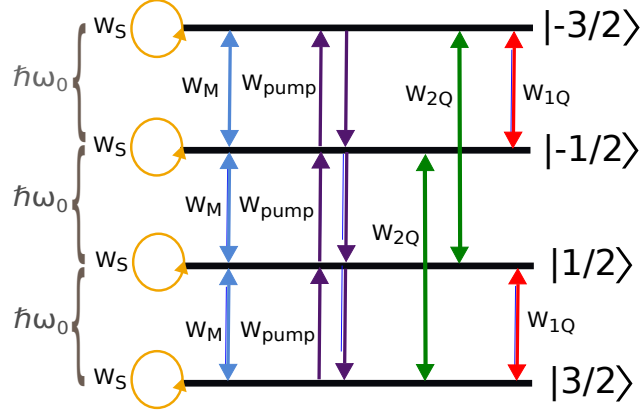


Figure 2.1: Environment-induced and the incoherent RF pump transitions in spin-3/2.

Hamiltonian when we discuss spin squeezing and the special advantages posed by the asymmetry term.

2.3 Four-Level System: Lindblad Operators

In this thesis the four-level system plays a particular role in connection to spin-3/2 nuclei like ^{69}Ga , ^{71}Ga , ^{75}As . In the presence of only an external magnetic field, the equally-spaced four states for a spin-3/2 nucleus are: $|3/2\rangle, |1/2\rangle, |-1/2\rangle, |-3/2\rangle$ shown in Figure 2.1. Since these aforementioned nuclei have positive gyromagnetic ratios, $|3/2\rangle$ becomes the ground state, and $|-3/2\rangle$ the highest excited spin state under a z -directed magnetic field.

Figure 2.1 also illustrates the environment-induced transition processes we shall be considering [15]. The associated transition rates being W_M , W_{2Q} , W_{1Q} , W_S .

- W_M is the magnetic dipole transition rate arising from the coupling to the fluctuating magnetic field at the site of the nuclear spin.
- W_{1Q} and W_{2Q} are respectively the single- and double-quantum transition rates associated with the coupling to the fluctuating EFG which only affect

the quadrupolar nuclei.

Note that the $1/2 \leftrightarrow -1/2$ so-called central transition is not affected by single-quantum quadrupole interaction [32, 33]. All these processes so far, directly change the state populations, and hence play decisive role in the so-called longitudinal or T_1 relaxation [36]. We need to consider also environment-induced processes that only cause phase decoherence without affecting the state populations. They govern the transverse or T_2 relaxation [36]. For this purpose we introduce the elastic dephasing or self-scattering with a rate W_S . In the context of quantum information theory, it is used for characterizing the phase-flip channel [37]. Finally, on this figure we also indicate transitions caused by an incoherent RF pump which will drive the system to saturation.

For spin-3/2 system, the corresponding Lindblad operators will be expressed by the SU(4) generalizations of the well-known Pauli spin matrices. The energy (Zeeman-term) operator which is also used in self-scattering has the form

$$\hat{I}_z \equiv \hat{I}_3 = \begin{bmatrix} 3/2 & 0 & 0 & 0 \\ 0 & 1/2 & 0 & 0 \\ 0 & 0 & -1/2 & 0 \\ 0 & 0 & 0 & -3/2 \end{bmatrix}. \quad (2.7)$$

The raising operator to be employed in magnetic transitions, and the quadrupolar

one- and two-quantum transition operators are

$$\begin{aligned}
\hat{I}_M^+ &= \begin{bmatrix} 0 & \sqrt{3} & 0 & 0 \\ 0 & 0 & 2 & 0 \\ 0 & 0 & 0 & \sqrt{3} \\ 0 & 0 & 0 & 0 \end{bmatrix}, \\
\hat{I}_{1Q}^+ &= \begin{bmatrix} 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 \end{bmatrix}, \\
\hat{I}_{2Q}^+ &= \begin{bmatrix} 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix}.
\end{aligned} \tag{2.8}$$

We obtain the spin-lowering operators (which cause increase in energy for the considered Zeeman term) by simply taking Hermitian conjugates of these raising operators

$$\begin{aligned}
\hat{I}_M^- &= (\hat{I}_M^+)^\dagger, \\
\hat{I}_{1Q}^- &= (\hat{I}_{1Q}^+)^\dagger, \\
\hat{I}_{2Q}^- &= (\hat{I}_{2Q}^+)^\dagger.
\end{aligned} \tag{2.9}$$

2.3.1 A Thermal Correction

When such a multi-level system reaches thermal equilibrium, the population should reduce to the Boltzmann distribution. That is, higher-lying states should be somewhat less populated $\rho_{00} > \rho_{11}$, which can be attained only when $W_- < W_+$; see, Figure 2.2. Essentially, this is due to the physical fact that there occurs both stimulated emission and absorption processes while there is only spontaneous *absorption* process [38]. Hence, down-in-energy electromagnetic transitions should always be stronger. Levitt has suggested to discriminate the up-and down-transitions rates as a simple means for thermal correction such

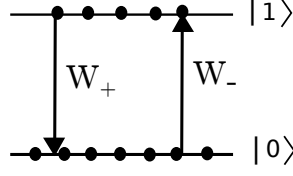


Figure 2.2: Thermal equilibrium state.

that

$$W_{\pm} \approx W(1 \pm \frac{1}{2}\beta), \quad (2.10)$$

where β is the Boltzmann factor which is $\frac{\hbar\gamma B^0}{k_B T}$ [39]. Here, $k_B T$ is the thermal energy and $\hbar\gamma B^0 \equiv \hbar\omega_0$ is the energy between Zeeman levels.

2.4 Coherent Spin State and Spin Squeezing

In this section, we are going to supply basic definitions and expressions for coherent spin states and spin squeezing. As an extensive and up-to-date resource, we refer to Ma et al.'s review paper [19].

2.4.1 Coherent Spin State (CSS)

We can generate a *coherent spin state* through rotating a Dicke state $|I, -I\rangle$ by azimuthal angle φ , and polar angle θ . Its mathematical form is given by [19]

$$|\theta, \varphi\rangle = \sum_{m=-I}^I \frac{1}{(1 + |\tau|^2)^I} \binom{2I}{m+I}^{1/2} \tau^{m+I} |I, m\rangle \quad (2.11)$$

where $\tau = e^{-i\varphi} \tan \frac{\theta}{2}$, and $\binom{n}{k}$ refers to binomial coefficient as in combinatorics.

2.4.2 Spin Squeezing

Spin squeezing is a means of reducing quantum fluctuation in one quadrature at the expense of the other quadrature so that overall Heisenberg uncertainty condition is not violated. In mathematical terms, for a spin vector having mutually orthogonal components I_i , I_j , I_k , while respecting

$$\langle \Delta I_i^2 \rangle \langle \Delta I_j^2 \rangle \geq \frac{1}{4} |\langle I_k \rangle|^2, \quad (2.12)$$

if either $\langle \Delta I_i^2 \rangle < \frac{1}{2} |\langle I_k \rangle|$, or (exclusively) $\langle \Delta I_j^2 \rangle < \frac{1}{2} |\langle I_k \rangle|$ then it corresponds to a spin-squeezed state.

To quantify the degree of squeezing, Kitagawa and Ueda [18] proposed the following squeezing parameter

$$\xi_S = \frac{(\Delta \hat{I}_n)_{min}}{\sqrt{I/2}}, \quad (2.13)$$

where, $(\Delta \hat{I}_n)_{min}^2$ is the smallest variance of the spin component $\hat{I}_n$ where $\hat{n}$ is perpendicular to $\langle \vec{I} \rangle$. $\xi_S = 1$ corresponds to CSS and a value below 1 indicates a squeezed state.

The smallest variance is calculated as [20]

$$(\Delta \hat{I}_n)_{min} = \sqrt{\frac{1}{2}C - \frac{1}{2}\sqrt{A^2 + B^2}}, \quad (2.14)$$

where, $A = \langle \hat{I}_z^2 - \hat{I}_y^2 \rangle$, $B = \langle \hat{I}_z \hat{I}_y + \hat{I}_y \hat{I}_z \rangle$ and $C = \langle \hat{I}_z^2 + \hat{I}_y^2 \rangle$. Here, the mean spin direction is chosen to be x axis, but in general, it needs to be determined separately.

Another relevant parameter is called *the squeezing angle* which is given by

$$\theta_{min} = \frac{1}{2} \tan^{-1}(B/A). \quad (2.15)$$

The squeezing angle shows how much the spin squeezing is tilted geometrically over the plane perpendicular to mean spin direction. When the squeezing angle is 0 we see *maximal squeezing*. In this situation, the squeezing parameter should

reach its minimum value in the period. This means, the squeezing parameter and squeezing are inversely proportional. Also, when the squeezing angle reaches its maximum positive value, the squeezing parameter also reaches its maximum value in that period.

Even though we shall be using Kitagawa's squeezing parameter ξ_S , we should mention that there are other estimates for squeezing [19], a popular variant being that introduced by Wineland et al.[40], related to ξ_S as

$$\xi_R = \frac{I}{|\langle \hat{I} \rangle|} \xi_S. \quad (2.16)$$

where, ξ_R is Wineland's squeezing parameter and $\langle \hat{I} \rangle$ is the mean spin. ξ_R is generally preferred in relating squeezing to entanglement which is a subject outside the scope of this thesis.

2.4.3 Generating Squeezing

In Kitagawa's original paper [18], two means of generating spin squeezing were proposed: the one-axis twisting Hamiltonian

$$\hat{H}_{OAT} = \chi \hat{I}_z^2, \quad (2.17)$$

and the two-axis countertwisting Hamiltonian

$$\hat{H}_{TAC} = \frac{\hbar\chi}{2i} \left(\hat{I}_+^2 - \hat{I}_-^2 \right). \quad (2.18)$$

It can be readily noticed that the quadrupole Hamiltonian in the presence of biaxility ($\eta \neq 0$) happens to be a combination of these two Hamiltonians, which we shall refer to within this context as mixed-axis twisting Hamiltonian [41]

$$H_{MAT} = \underbrace{\frac{e^2 q Q}{4I(2I-1)}}_{hA_Q} \left[3\hat{I}_z^2 - \hat{I}^2 + \eta \frac{\hat{I}_+^2 + \hat{I}_-^2}{2} \right]. \quad (2.19)$$

2.4.4 Quasi Probability Distributions

To understand how the spin squeezing changes and oscillates between the maximum and minimum values, the Quasi Probability Distribution (QPD) on the Bloch sphere was used in the original papers [20], [22]. The QPD helps us to see the uncertainty of a spin and how the direction of spin squeezing changes in time.

In the paper by Jin and Kim [20] the Husimi Q function is used to plot QPD

$$Q(\theta, \phi) = |\langle \theta, \phi | \Psi(t) \rangle|^2. \quad (2.20)$$

Here, the general form of coherent spin state is used as $|\theta, \phi\rangle = \exp\left\{-i\theta(\hat{I}_x \sin \phi - \hat{I}_y \cos \phi)\right\} |I, -I\rangle$. The spin dynamics under quadrupolar Hamiltonian, using Wigner and Husimi functions can deceptively give the same visual distribution. In general while Husimi function is always positive, Wigner function can have negative values in the distribution [38].

Additional to QPD distribution, calculating the probability amplitudes of each spin projection gives the means to determine the variance of the squeezing [20]. To do this, we define a state vector

$$|\psi(t)\rangle = \sum_m C_m(t) |I, m\rangle \quad (2.21)$$

where $-I \leq m \leq I$. Solving the time dependent Schrödinger Equation, we can calculate how the squeezing increases or decreases by monitoring $C_m(t)$.

Chapter 3

Saturation and Relaxation in Spin-3/2 Nuclei

In this chapter, we study how a spin-3/2 nucleus is driven to saturation under an incoherent radio frequency (RF) pumping, and subsequently how it relaxes back to equilibrium when left to its own devices. In particular, we search for the measurable traces of electric quadrupolar interaction in either of these processes.

3.1 General Description of the System

Let's recall the form of the Lindblad master equation from the previous chapter.

$$\frac{d}{dt}\hat{\rho}_S(t) = -i \left[\hat{H}, \hat{\rho}_S(t) \right] - \frac{1}{2} \sum_{\mu} \kappa_{\mu} \left\{ \hat{\mathcal{L}}_{\mu}^{\dagger} \hat{\mathcal{L}}_{\mu} \hat{\rho}_S(t) + \hat{\rho}_S \hat{\mathcal{L}}_{\mu}^{\dagger} \hat{\mathcal{L}}_{\mu} - 2 \hat{\mathcal{L}}_{\mu} \hat{\rho}_S(t) \hat{\mathcal{L}}_{\mu}^{\dagger} \right\} , \quad (3.1)$$

Throughout this chapter the Hamiltonian simply includes the external magnetic field,

$$\hat{H} = -\hbar\omega_0 \hat{I}_z . \quad (3.2)$$

where ω_0 is the Larmor angular frequency. In the following chapter we shall also

add the quadrupole interaction directly to the Hamiltonian. For now, it will only be considered though environmental fluctuations.

The list of collapse operators $\{\hat{\mathcal{L}}_\mu\}$ are as follows: an incoherent but resonant RF drive as is commonly used in NMR, and magnetic fluctuations in the environment (a lumped sum of both vacuum field and the thermal fluctuations) that trigger magnetic dipole transitions. For these, we make use of the lowering and raising operators $\hat{I}_{-/+}$; the associated rates will denoted by W_{pump} , W_M . In the most general case, we also account for quadrupolar interaction with the environment via raising and lowering operators of single-quantum $\hat{I}_{1Q}^+$ and $\hat{I}_{1Q}^-$, and double-quantum transitions $\hat{I}_{2Q}^+$ and $\hat{I}_{2Q}^-$, with rates W_{1Q} and W_{2Q} . Finally, the elastic dephasing (sometimes termed as self-scattering) is represented by the operator I_z with a rate W_S . Other than the RF pump, the up- and down-transition rates are also subjected to thermal correction according to Eq. (2.10).

3.2 Reproducing The Experimental T_1 and T_2

To determine the rate values one needs to consider some relevant experiments. We make use of the recently published data by Yesinowski on ^{71}Ga nuclei [15] $T_M = 50$ s, $T_{1Q} = 3$ s and $T_{2Q} = 3$ s. However, this data is only useful to set the diagonal (i.e., longitudinal) relaxation time. For the off-diagonal (i.e, transverse) relaxation time, Chekhovich et al. measured at cryogenic temperatures a typical value of 4 ms for InGaAs quantum dot nuclei [42] where the spin value of In is 9/2 while Ga and As have the spin value 3/2. To fit the latter, we include the aforementioned self-scattering process.

To be able to reproduce these experimental values, we set the rates as

$$\begin{aligned} W_M &= 6.96 \text{ mHz}, \\ W_{1Q} &= 327.12 \text{ mHz}, \\ W_{2Q} &= W_{2Q}, \\ W_S &= 515.41 \text{ Hz}. \end{aligned} \tag{3.3}$$

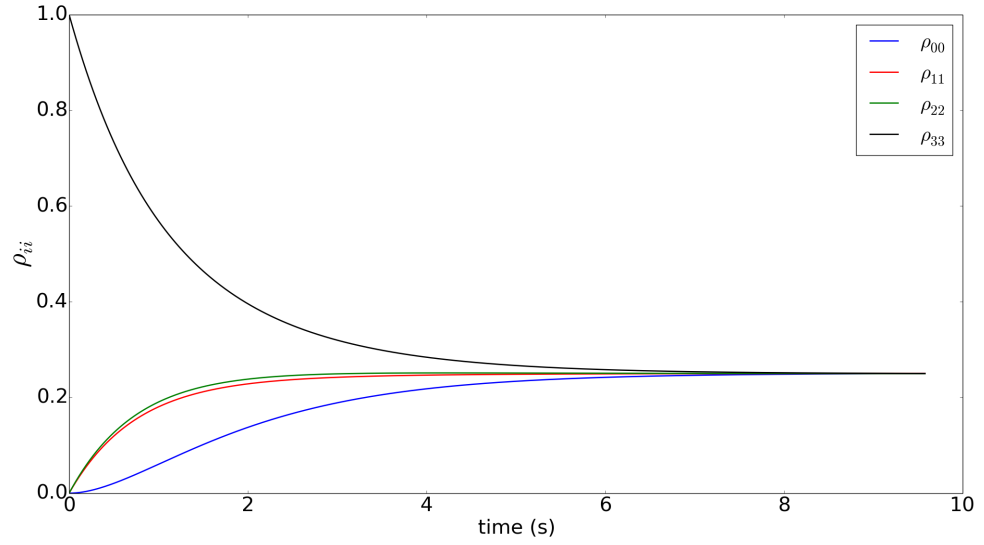


Figure 3.1: Diagonal terms under realistic collapse rates.

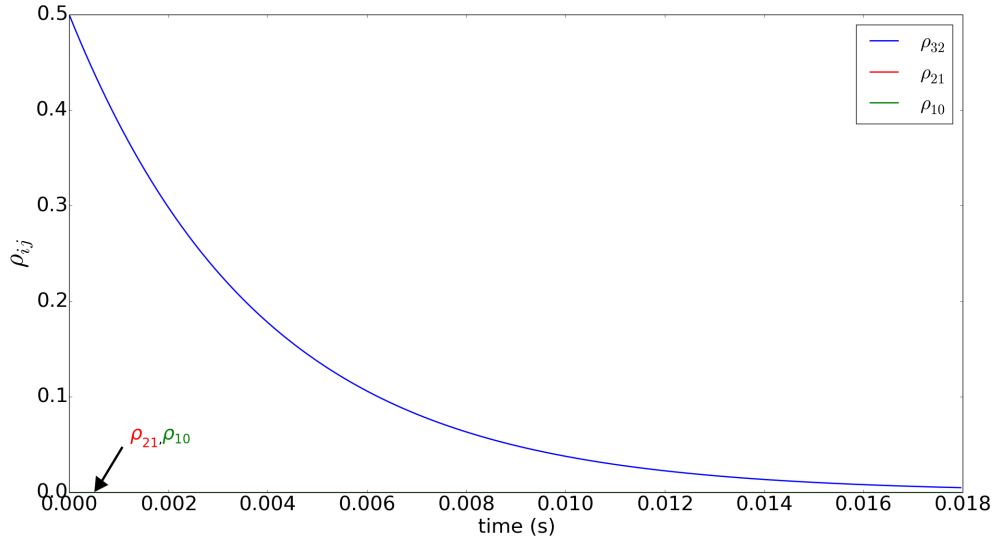


Figure 3.2: Off-diagonal terms under realistic collapse rates.

Under this model, and without any RF pump we let the system start from the initial state

$$|\psi(t = 0)\rangle = |3\rangle \quad (3.4)$$

and reach thermal equilibrium. The evolution of the diagonal part of the density matrix can be seen in Figure 3.1. For monitoring the off-diagonal dynamics, we start from an initial state

$$|\psi(t = 0)\rangle = \frac{1}{\sqrt{2}} (|2\rangle + |3\rangle) \quad (3.5)$$

and, as shown in Figure 3.2 this coherent state decays when the system attains equilibrium. Note that the other off-diagonal entries are not visible in this graph in this scale.

From these two plots we can extract T_1 and T_2 values as

- for diagonal elements: $T_1 = 1.504$ s,
- for off-diagonal elements: $T_2 = 3.866$ ms.

Note that these were the intended experimental values mentioned above [15], [42].

3.3 Saturation and Relaxation

Next, we start from an initial thermal state

$$\rho(t = 0) = \rho_{thermal} , \quad (3.6)$$

which is governed by $\frac{\hbar\omega_0}{k_B T} = 0.005$. Here, we envision the case of 10 T magnetic field, for the spin-3/2 nuclei such as ^{75}As , ^{69}Ga , ^{71}Ga , having a typical gyromagnetic ratio of 10 MHz/T value under liquid helium temperatures.

Now, we turn on an incoherent RF pump with a rate W_{pump} ; when we increase it, we expect to see faster saturation the of the level populations. We set the

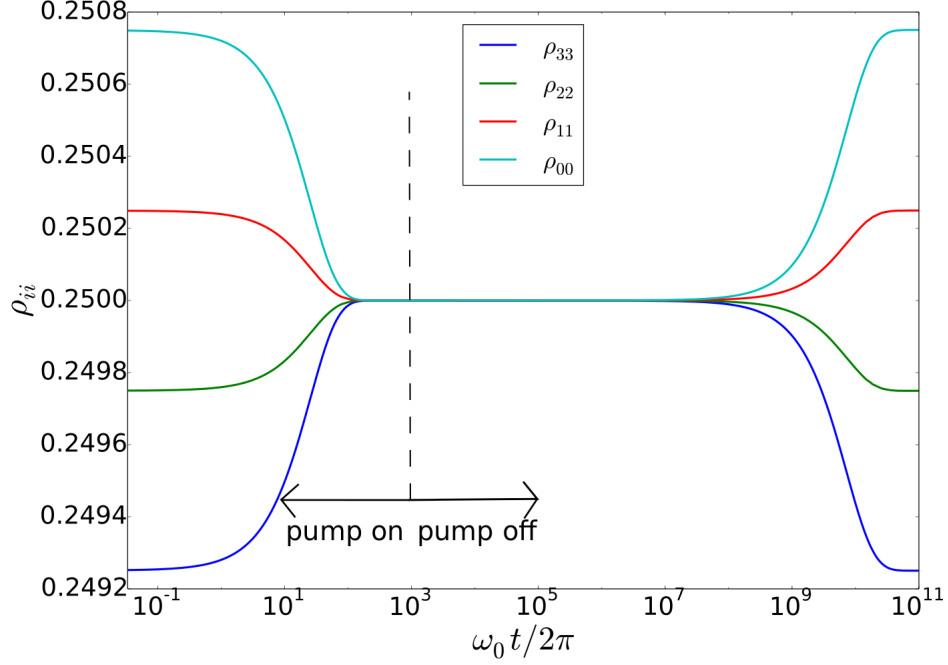


Figure 3.3: Saturation (RF pump on) and subsequent relaxation (RF pump off) of spin-3/2 system starting from the thermal state without quadrupolar transitions.

pump rate in relation to the linear Larmor frequency f_0 as

$$\begin{aligned} f_0 &= 100 \text{ MHz}, \\ W_{\text{pump}} &= f_0/50. \end{aligned} \tag{3.7}$$

After the system saturates, we turn off the pump to monitor how it recovers the thermal state under the action of all collapse operators with the exception of quadrupole operators, as shown in Figure 3.3.

In Figure 3.4 we illustrate the case when the quadrupolar transitions are also included. The only distinction with respect to Figure 3.3 is that the recovery from saturation occurs much faster due to added channel of quadrupolar transitions.

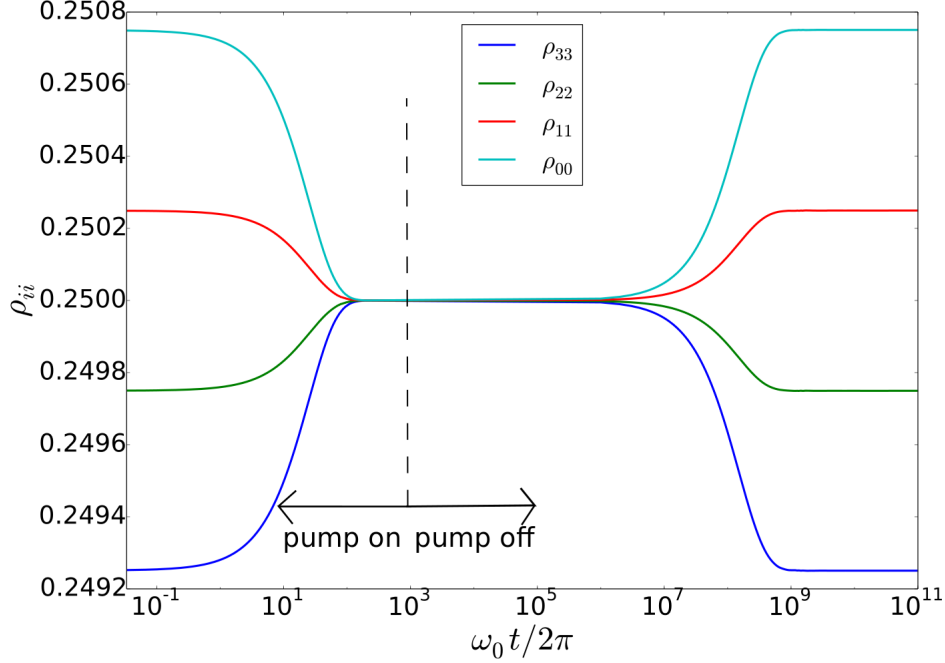


Figure 3.4: Saturation (RF pump on) and subsequent relaxation (RF pump off) of spin-3/2 system starting from the thermal state with quadrupolar transitions.

3.3.1 Loss of Coherence

So far, we only dealt with the effect of environment on the state populations, that are diagonal components of the density matrix. In order to probe how coherences, in other words the off-diagonal entries of the density matrix are affected by these processes, we initialize the system to a superposition of states, as given by

$$|\psi(t=0)\rangle = \frac{1}{\sqrt{2}} (|0\rangle + i|1\rangle + |2\rangle + i|3\rangle) \quad (3.8)$$

where $\hat{\rho}(t=0) = |\psi(t=0)\rangle \langle\psi(t=0)|$.

For this, we keep the parameters that we used in previous sections, however we do not pump the system, but solely let it relax to the thermal state. The transitions also include the quadrupolar transitions. The evolution for real parts of the elements of density matrix is shown in Figure 3.5, and the imaginary part in Figure 3.6 at several time instants indicated in units of the Larmor period T_0 . The loss of coherence can be easily seen through the imaginary parts. In real

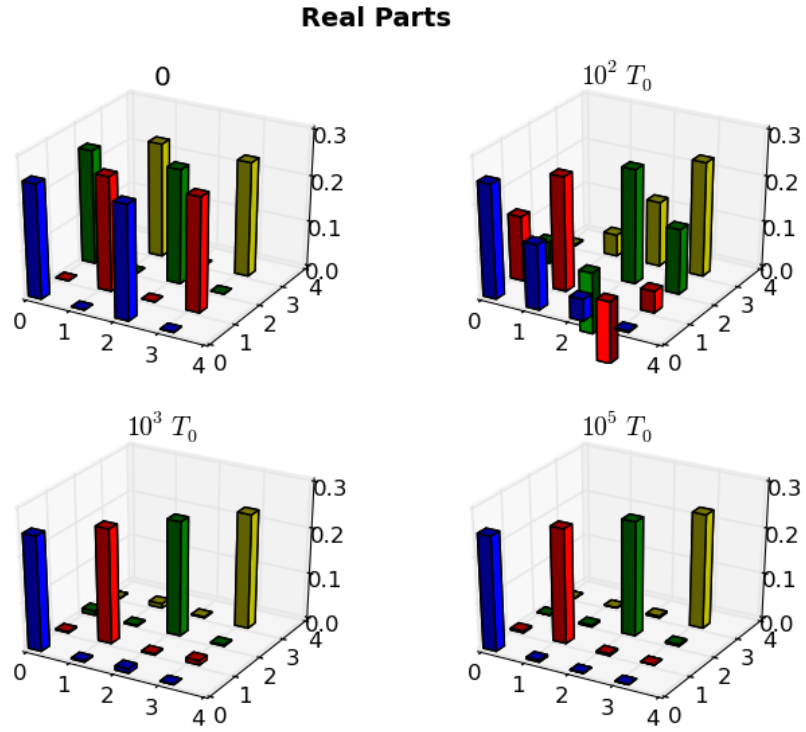


Figure 3.5: Real part of the results starting from the superposition of states.

part, we see first the system has both populations and coherences, but in the end because the system reaches its equilibrium state, we only see the thermal populations as expected from our previous results.

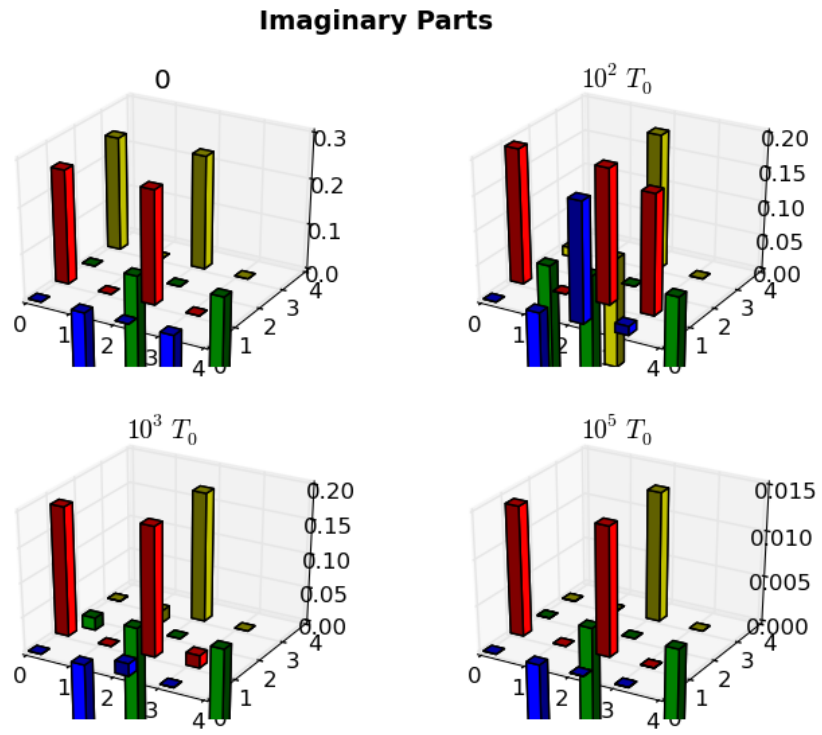


Figure 3.6: Imaginary part of the results starting from the superposition of states.

Chapter 4

Nuclear Spin Squeezing

In this chapter, we study spin squeezing with one-axis twisting (OAT), two-axis countertwisting (TAC), mixed-axis twisting (MAT) which is the intrinsic form of electric quadrupole interaction. The inspiration for this chapter comes from the two papers [22] [20] that we have mentioned in Chapter 2. As we are interested in quadrupolar nuclei, we only consider $I = 1, \dots, 9/2$ spins in this chapter.

4.1 OAT, TAC and MAT

In this section, we investigate the squeezing characteristics of spin values $I = 1 \dots 9/2$ starting from a CSS initial state. In Figure 4.1 we plot the Kitagawa squeezing parameter ξ_S for these spin values under OAT, TAC and MAT. For the latter, we choose an EFG asymmetry value of $\eta = 0.5$. We use a normalized time $A_Q t$, where A_Q is the quadrupole frequency as indicated in Eq. 2.19. The common trend is that as the spin value increases while OAT and TAC periodically oscillate in a band bounded by unity from above, the MAT varies over a broader range exceeding the unity squeezing value of a CSS; note that $\xi_S > 1$ is indeed possible [19]. Furthermore, unlike OAT and TAC, MAT displays a quasi-periodic pattern.

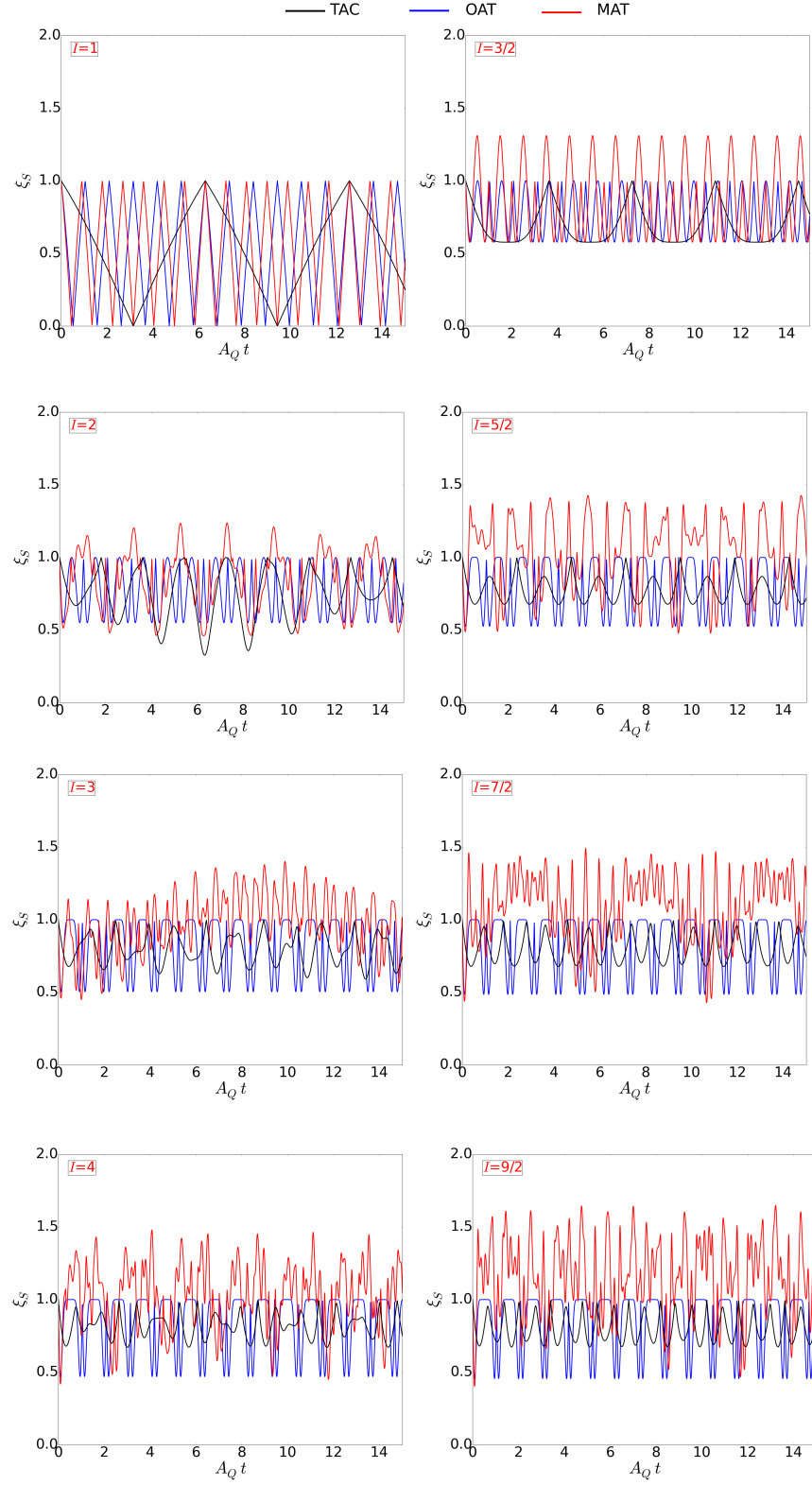


Figure 4.1: Kitagawa squeezing parameter ξ_S for values $I = 1, \dots, 9/2$ under OAT, TAC and MAT. For the latter $\eta = 0.5$.

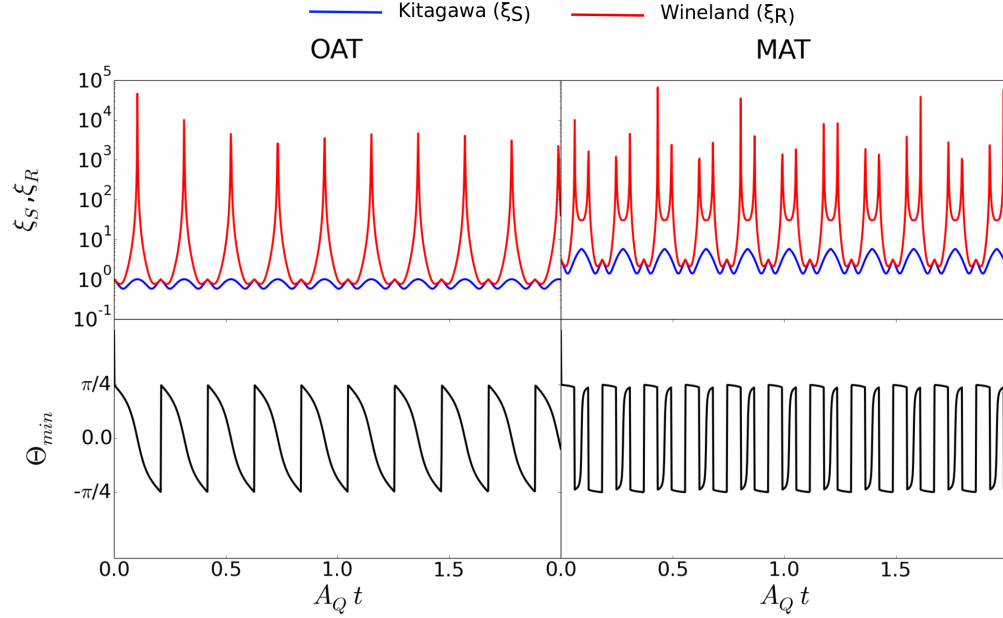


Figure 4.2: The ξ_S , ξ_R and θ_{min} for $I = 3/2$ under OAT and MAT ($\eta = 0.5$).

4.1.1 Kitagawa vs. Wineland

In Figure 4.2, we see that Wineland squeezing parameter ξ_R can reach much higher values than Kitagawa's squeezing parameter. Even though the amplitude changes, they oscillate in the same periodicity. The Wineland parameter is preferred in the context of entanglement among many $1/2$ spins that make up a Bose-Einstein condensate [19], which is out of the scope of our work. From the lower panel of the same plot we can see that the squeezing angle has more rapid oscillations under MAT.

4.1.2 Quasi Probability Distributions

To shed more light on the squeezing fluctuations, we compare in Figure 4.3 the OAT and MAT QPD at four different time instants within a period for $I = 9/2$, using the Husimi-Q function and the probability distributions of spin projections. Here, the light blue colors represent the minimum uncertainty quadrature, and the dark blue bar plots correspond to the maximum uncertainty quadrature.

In the case of CSS (as in $t=0$ instants) the uncertainty of the two quadratures become equal, yielding $\xi_S=1$.

4.2 Squeezing With Different η Values

To see how the squeezing changes with different EFG asymmetry parameter, η values, we plot $\eta=0, 0.1, 0.3, 0.5, 0.7, 1$ for $I=3/2$ and $9/2$ in Figure 4.4 and 4.5. For $\eta = 0$, OAT and MAT trivially coincide. When η increases we clearly observe that the initial CSS shifts predominantly towards anti-squeezed zone, while the OAT curves remain squeezed. This pattern is enhanced for higher spins, as can be seen from $I=9/2$. The main conclusion to be drawn from this analysis is that the bare quadrupole Hamiltonian that generates MAT inherently drives the nuclear spins to anti-squeezed behavior. In other words the quadrupole interaction can adversely affect the taming of the nuclear spin bath.

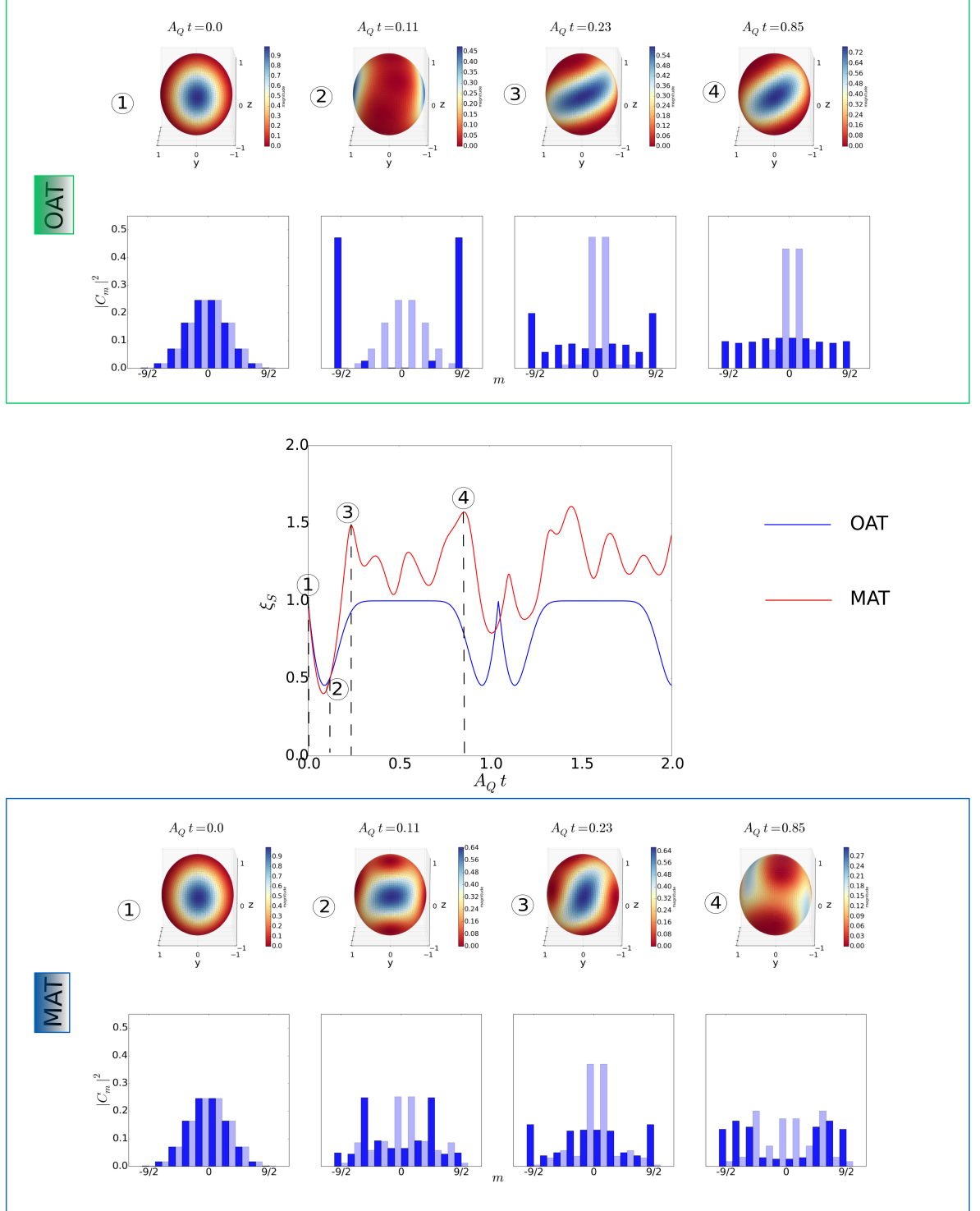


Figure 4.3: QPD for $I = 9/2$ under OAT and MAT ($\eta = 0.5$).

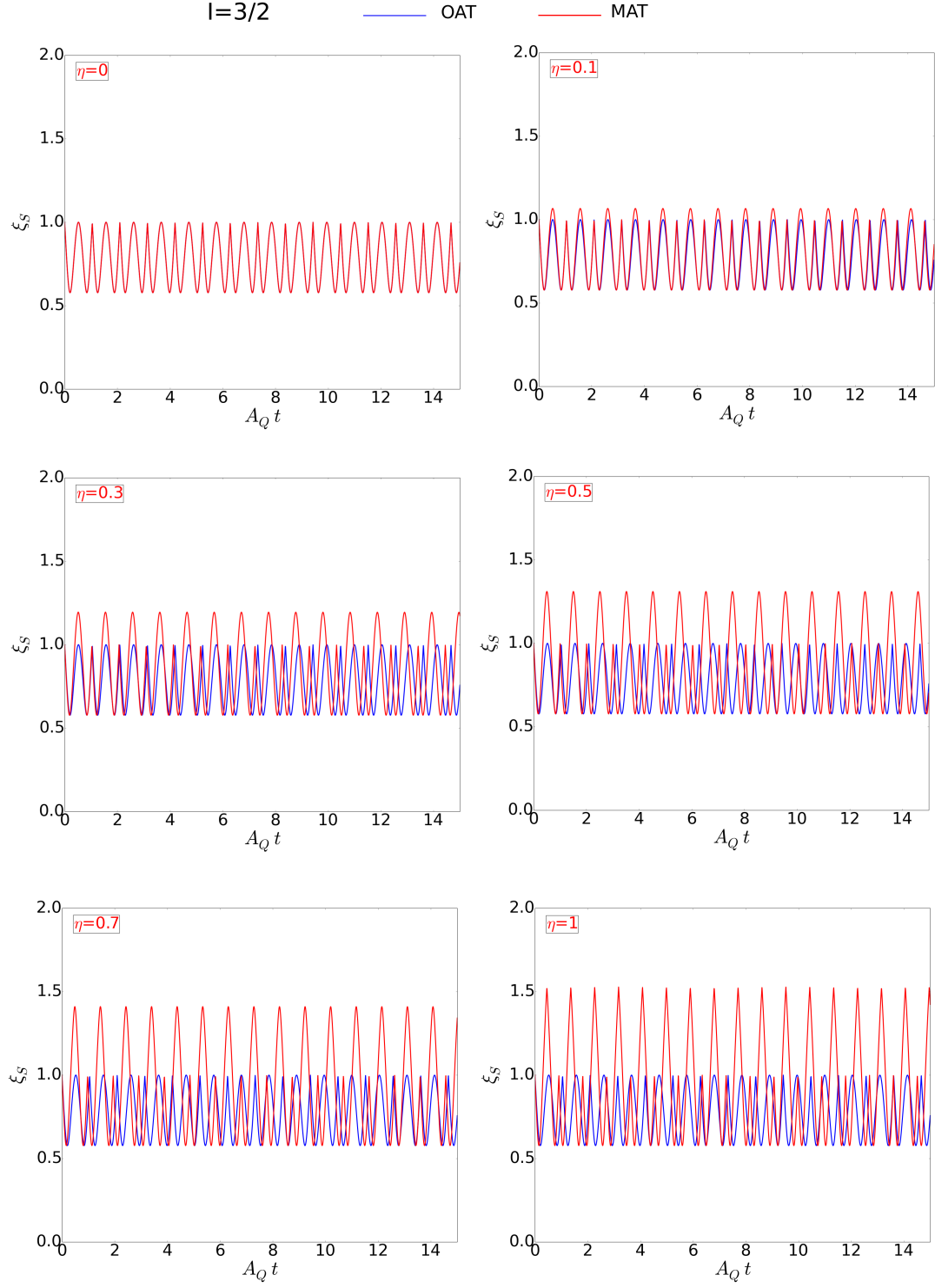


Figure 4.4: Squeezing parameter for $I = 3/2$ with different η values.

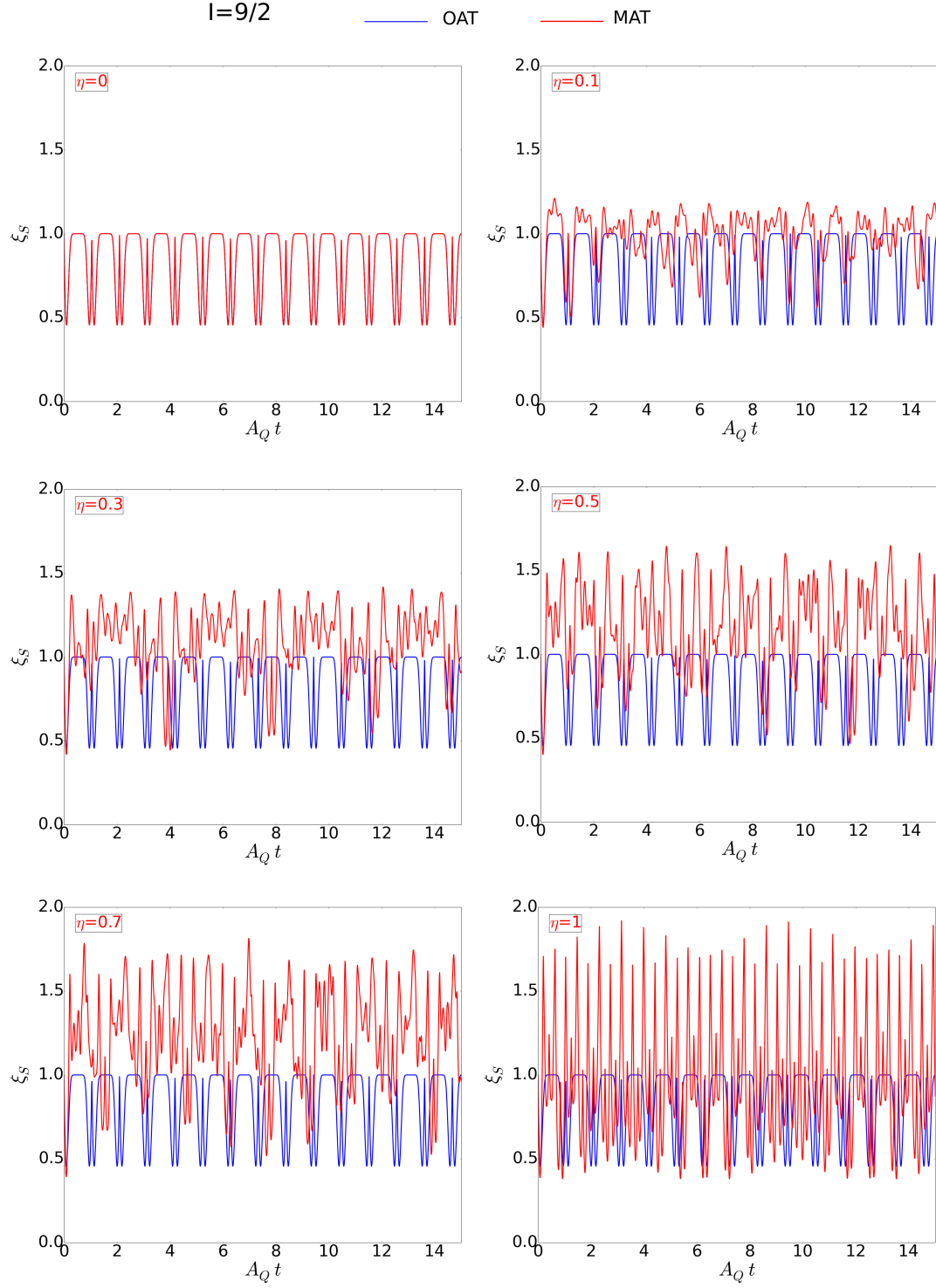


Figure 4.5: Squeezing parameter for $I = 9/2$ with different η values.

Chapter 5

Conclusions

In this thesis, we focused on two different kind of problems for nuclear spins involving quadrupole interaction. The first one that we examine in Chapter 3, is the saturation and relaxation process of $3/2$ -spins. As the second problem, in Chapter 4 we tried to understand the effect of spin squeezing via quadrupole interaction for spins $I = 1, \dots, 9/2$.

In the first part, we used the Lindblad master equation formalism with a system Hamiltonian involving only the external magnetic field, in contact to an incoherent RF pump, magnetic dipole and electric quadrupolar as well as elastic dephasing fluctuations of the environment. First, we extracted the rates of the out-of-system couplings from recent experiments. The longitudinal T_1 , and transverse T_2 relaxation times have been reproduced by monitoring the decay of the diagonal and off-diagonal entries of the density matrix, respectively. Using different initial states for the spins, and with and without including the RF pumping, we examined the population saturation and relaxation, and the loss of coherence. For these type of experiments, the sole effect of quadrupole interaction is to cause somewhat faster reach to the steady-state; be it saturation under RF pump, or relaxation in the lack of it.

In the second part, we considered a Hamiltonian involving only the quadrupole

interaction, and investigated how does the squeezing of an initial CSS evolve in time under limiting EFG asymmetry values η that lead to OAT, TAC and MAT for spin values $I = 1 \dots 9/2$. Moreover, we plotted Husimi QPD and quadrature amplitude distributions to aid the visualization of the changes in squeezing. The common trend is that MAT, which is the native Hamiltonian of a nuclear quadrupolar spin, leads to anti-squeezing especially for higher spins. This conclusion is further substantiated by analyzing the effect of increasing η for $I = 3/2$ and $9/2$ cases. This can be interpreted as quadrupole interaction having adverse effects against quieting the nuclear spin bath.

For future work, we would like to improve our rudimentary understanding of squeezing by adding other terms in the Hamiltonian like a coherent RF pulse that can rotate spins over the Bloch sphere, axial static magnetic field, as well as classical noise to see how squeezing of the system can be manipulated through these "control knobs. Furthermore, we want to combine the relaxation part with the squeezing part and extend the discussion of the latter to an open spin system.

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