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Magnetic Resonance Force Microscopy and the spin bath : towards single-spin massive-resonator entanglement and the spoiling influence of the spin bath

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1 *Introduction*

BUILDING EFFECTIVE RELATIONSHIPS requires speaking the same language. It is remarkable that although we formulate physics in the language of mathematics, nature itself speaks it better. After enormous investments into this relationship, physicists started seeing two different natures of nature. One of them is elegantly described by Einstein's geometric theory of gravitation,¹ better known as general relativity. The other, the nature of small things, is best described by a theory of complex valued probability amplitudes: quantum theory.

The unfeasible unification of the two theories describing both natures is one of the biggest conundrums humanity ever faced. The solution is of utmost importance as it might reveal where the universe's existence originates from, but also whether the world is deterministic or not.² Despite incredible efforts in the last century, physicist and mathematicians did not succeed creating a consistent theory of everything, yet.

A large problem of quantum theory is the understanding of why the squared norm of the normalized quantum states gives a probability distribution that describes the possible measurement outcomes, also known as Born's rule. There are several interpretations to overcome this measurement problem. In my opinion, the many worlds interpretation³ has a great resemblance with the worldview of the prisoners in Plato's cave. To free ourselves from the cave we need to ask nature itself for more information. However, performing measurements beyond quantum mechanics is a difficult

¹ Einstein 1915

² A subject closely related to free will, see Atmanspacher 2015.

³ This interpretation takes quantum theory for granted, and avoids the measurement problem.

thing to do. Luckily we are on the edge of a new era of technical possibilities where we can push systems over the supposed safe boundaries of quantum theory. We then have to compare the outcomes of these measurements with the conventional quantum theory and the different interpretations, and other beyond quantum mechanics theories.⁴

⁴ Bassi et al. 2013

1.1 *Spin mechanics*

MANY RESEARCH GROUPS that are exploring the boundaries of quantum mechanics are trying to find a non-classical state of a mechanical object⁵ due to the interaction with an easily controllable quantum state (qubit). A popular version is a resonating mirror that is part of a cavity for photons. The branch of physics studying this system is called cavity optomechanics. The cavity can be replaced with other qubit-holding systems which, together with the mechanical object, can be called a hybrid quantum system.

⁵ Here ‘mechanical object’ means a mass with something attached, such as a mirror, a magnet, or just being conductive, such that it can interact with some physical field (usually the electromagnetic field).

IN THIS THESIS we describe and work towards an experiment that should eventually be useful in verifying/falsifying gravitational induced spontaneous collapse models³ such as the Diósi-Penrose model⁶ and closely related models.^{7,8} The basic idea, that widespread⁹ wave functions are energetically unfavorable for the gravitational field¹⁰ compared to collapsed wave functions, can be tested by creating larger and larger position-separated superpositions of macroscopic objects. These superpositions can be created by coupling a well controlled quantum object to the macroscopic one. The force (or interaction strength) a single qubit can exert onto the mechanical object is limited and therefore a low spring constant is necessary to create a large position displacement of the mass.¹¹ The setup we choose to develop is a Magnetic Resonance Force Microscope (MRFM) coupled to a Nitrogen-Vacancy center¹² (NV⁻-center, or just NV) for several reasons: First because MRFM is a technique where the basics

⁶ Penrose 2014

⁷ Oosterkamp and Zaanen 2013

⁸ Rademaker et al. 2014

⁹ Widespread as function of position.

¹⁰ Or better said: spacetime.

¹¹ More on this in Ch. 5

¹² Doherty et al. 2013

have been developed in the last two decades, and nowadays there are ultrasoft cantilevers available with spring constants less than 50 $\mu\text{N/m}$. Moreover, there is a whole range of spin manipulation protocols created that can directly be used.¹³ The qubit connected to our mechanical object in MRFM is a spin which is a big advantage as spin-qubits can decay and decohere very slowly. Although a nuclear spin is much more stable than an electron spin, the latter has a larger magnetic moment by three orders of magnitude and therefore a larger interaction strength by the same amount. NV-centers show the longest longitudinal (T_1) and transversal (T_2) decay times of individual electron-spin like spins¹⁴ and we argue in Ch. 5 that this is enough for our experiment. The biggest advantage of NV-centers is that they can be very precisely controlled using light and radio-frequent (RF) fields.¹² Finally it should be noted that creating a hybrid quantum system in this way, also contributes to the development of the MRFM technique, thereby making it a win-win situation. Even when in a follow-up research it turns out that the developed experiment becomes too difficult or does not give the results one could have hoped for, it most definitely has been useful for developing and commercializing the MRFM, and has provided new important single atom analysis methods to condensed matter scientists,¹⁵ biophysicists,¹⁶ and probably various industries.

¹³ Poggio and Degen 2010

¹⁴ Bar-Gill et al. 2013

¹⁵ Wagenaar et al. 2016

¹⁶ Degen et al. 2009a

BESIDE VERIFYING THEORIES, experimental results can also point towards a yet unknown theory, such as happened after the famous Michelson-Morley experiment, and the Stern-Gerlach experiment. On a smaller scale we have also seen this in this thesis: the temperature dependent dissipation experiment described in Ch. 3 helped finding the general theoretical results of Ch. 2 where we explain how a paramagnetic spin can significantly influence the resonance frequency and dissipation of a macroscopic resonator. We verified this theory and used it for the new experiment (Ch. 4-5).

A PRECISE UNDERSTANDING OF RESONATORS is necessary for all chapters in this thesis. Therefore, we continue this chapter by summarizing the basics regarding classical mechanical resonators without any specific interaction with other systems. The same principles apply to electromagnetic resonators, but as we only need this in Sec. 2.4 it is left aside for the moment. In Sec. 1.2 we provide a Lagrangian description of a bare mechanical resonator, in Sec. 1.3 we give a treatment of the thermal motion, while in Sec. 1.4 we calculate under which conditions driving the resonator may heat the system it is coupled to. In Sec. 1.5 we describe the contents of the chapters in this thesis and how these are related to each other.

1.2 Mechanical resonators

THE CLASSICAL MOTION OF THE CANTILEVER can be determined by minimizing the action. For small displacements q the cantilever can be thought of as a harmonic oscillator, whose Lagrangian, L , is

$$L = T - V = \frac{1}{2}m\dot{q}^2 - \frac{1}{2}k_0q^2. \quad (1.1)$$

Here, T and V are the kinetic and potential energy, respectively. q and $\dot{q}$ are the generalized coordinates of the position and velocity respectively. Furthermore, m is the effective mass of the cantilever, k_0 the spring constant which the cantilever would have in case there is no interaction with parts outside the system.

As for small displacements q can be taken to point in a single Cartesian direction, we can work with the scalar q .¹⁷ Let us continue finding a classical solution for q . Minimizing the action gives us the equations of motion (EOM)

$$\frac{d}{dt} \frac{\partial L}{\partial \dot{q}} - \frac{\partial L}{\partial q} = F_{\text{ext}}(t) \Rightarrow m\ddot{q} + k_0q = F_{\text{ext}}(t). \quad (1.2)$$

The external force term is added manually based on the second law of Newton. Note that with F_{ext} the energy in the

¹⁷ Note that q can also be negative, so not q , but $|q|$ is the norm of q .

system is not conserved. In fact we should also take the dissipative mechanisms into account since this makes the cantilever move differently. The dissipative force for a harmonic resonator can be thought of as a viscous drag $-\gamma\dot{q}$ because of the movement and friction in the spring and surroundings. Since the dissipated energy depends on the path the cantilever takes, the force cannot be derived from a potential description. Therefore we manually add this dissipation term as a special kind of an external force. Rewriting the EOM gives

$$F_{\text{ext}}(t) + F_{\text{fric}}(\dot{q}) - \frac{\partial V}{\partial q} = \frac{d}{dt} \frac{\partial T}{\partial \dot{q}} - \frac{\partial T}{\partial q}, \quad (1.3)$$

where the right-hand side would be a conserved quantity (the generalized force) in case the cantilever system would be dissipationless and not influenced from the outside. From the last equation it is easy to see that we can effectively recreate a conserved system by choosing $F_{\text{ext}}(t) = -F_{\text{fric}}(\dot{q})$. We want this so we are able to do continuous measurements, but therefore we should know the path $\dot{q}$ first.

IF WE FILL IN the terms of the last equations we find

$$m\ddot{q} + \gamma\dot{q} + k_0q = F_{\text{ext}}(t). \quad (1.4)$$

The solution of this inhomogeneous ordinary differential equation can be found in several ways. We use the Laplace transform, $\mathcal{L}\{q\}(s) = \int_0^\infty q(t)e^{-st}dt$, as we need to use that as well in Ch. 2. If we shift the time-axis such that an arbitrarily chosen initial time $t_0 \rightarrow 0$, we find

$$\begin{aligned} \mathcal{L}\{q\}(s) = & \frac{\left(s - \frac{\omega_0}{2Q}\right)q(0) + \omega_r \left(\frac{q(0)}{\sqrt{4Q^2-1}} + \frac{\dot{q}(0)}{\omega_r}\right)}{\left(s + \frac{\omega_0}{2Q}\right)^2 + \omega_r^2} \\ & + \frac{1}{\omega_0^2 + s^2 + \frac{\omega_0}{Q}s} \frac{\mathcal{L}\{F_{\text{ext}}\}(s)}{m}, \end{aligned} \quad (1.5)$$

where ω_0 is the natural frequency $\sqrt{\frac{k_0}{m}}$, Q the quality factor $\frac{\sqrt{k_0m}}{\gamma}$, and $\omega_r \equiv \pm\omega_0\sqrt{1 - \frac{1}{4Q^2}}$, which is the frequency

where the resonance is the strongest. $\mathcal{L}\{q\}(s)$ has two parts: the transients and the steady state. The transient solution is found by setting $F_{ext} = 0$, so it only depends on the initial conditions $q(0)$ and $\dot{q}(0)$. Returning to the time-domain¹⁸ we find

$$q(t) = e^{-\frac{\omega_0}{2Q}t} \left[q(0) \cos(\omega_r t) + \left(\frac{q(0)}{\sqrt{4Q^2 - 1}} + \frac{\dot{q}(0)}{\omega_r} \right) \sin(\omega_r t) \right]. \quad (1.6)$$

This solution will always decrease exponentially to the solution $q = 0$ and therefore it will be of no interest for continuous experiments. However, a so called ring down experiment, where one measures the response after giving the resonator a certain $q(0)$ or $\dot{q}(0)$, is an efficient way to measure ω_r and Q for resonators with large Q -factors.

THE STEADY STATE SOLUTION does not depend on the initial conditions and is of much more interest for us as we would like to use the resonator as a continuous detector. The steady state solution is basically the last part of Eq. 1.5. When we drive the system¹⁹ we can represent the system in the frequency domain²⁰ and find

$$\tilde{q}(\omega) = \frac{e^{i\phi(\omega)}}{\sqrt{(\omega_0^2 - \omega^2)^2 + \left(\frac{\omega_0\omega}{Q}\right)^2}} \frac{\tilde{F}(\omega)}{m}, \quad (1.7)$$

where the phase ϕ can be calculated using the four-quadrant inverse tangent²¹ $\phi(\omega) = \text{atan2}\left(\frac{-\omega_0\omega}{Q}, \omega_0^2 - \omega^2\right)$.

For a sinusoidal force $F_{ext} = F_0 \sin(\omega_d t)$ the response of the system is

$$q(t) = A(\omega_d) \sin(\omega_d t + \phi(\omega_d)), \quad \text{with amplitude} \quad (1.8)$$

$$A(\omega) \equiv \frac{\frac{F_0}{m}}{\sqrt{(\omega_0^2 - \omega^2)^2 + \left(\frac{\omega_0\omega}{Q}\right)^2}}.$$

¹⁸ Technically we should assume $\text{Re}\{s\} > -\frac{\omega_0}{2Q}$. This is automatically satisfied as for the transients we are only interested in $\text{Re}\{s\} = 0$ (the frequency domain).

¹⁹ Just driving the resonator without feedback turned on.

²⁰ As there are no poles on the imaginary axis of s we can take $s \rightarrow i\omega$, $\mathcal{L}\{q\}(s) \rightarrow \tilde{q}(\omega)$, and $\mathcal{L}\{F_{ext}\}(s) \rightarrow \tilde{F}(\omega)$.

²¹ The range of $\text{atan2}(y, x)$ is $(-\pi, \pi]$, rather than $(-\frac{\pi}{2}, \frac{\pi}{2})$ for $\text{atan}(\frac{y}{x})$.

1.3 Thermal noise

FROM THE FLUCTUATION DISSIPATION THEOREM it follows that the thermal force noise is related to the imaginary part of the Fourier transform of the linear response function of the force, i.e. the force $\tilde{F}(\omega)$ that the cantilever feels when it is moved by $q(\omega)$. It follows that the one-sided spectral density function of the force noise is given by

$$S_F(\omega) = \frac{4k_B T}{\omega} \text{Im} \left(\frac{\tilde{F}(\omega)}{q(\omega)} \right) = 4k_B T \gamma. \quad (1.9)$$

The (also one-sided) spectral density function of the position of the cantilever is then found by substituting $S_F(\omega)$ into the expression for $q(\omega)q(\omega)^*$ which gives

$$S_q(\omega) = \frac{4k_B T}{m} \frac{\frac{\omega_0}{Q}}{(\omega_0^2 - \omega^2)^2 + \left(\frac{\omega_0 \omega}{Q}\right)^2} \quad (1.10)$$

$$\approx \frac{k_B T}{k_0} \frac{\frac{\omega_0}{Q}}{(\omega_0 - \omega)^2 + \left(\frac{\omega_0}{2Q}\right)^2}. \quad (1.11)$$

In the last step we approximated the result by a Lorentzian distribution. This can be done by expanding the denominator until second order in $\omega_0 - \omega$, and neglecting higher order terms and the term $\omega_0^2 \frac{\omega_0^2 - \omega^2}{Q^2}$. This approximation is thus only valid for high Q and near resonance $\omega \approx \omega_0$. However, for our experiments the error is neglectable.²² For Lorentzian distributions it can be shown that the full width at half maximum (FWHM) is $\frac{\omega_0}{Q}$.

²² For example consider a resonator with its resonance frequency at 3 kHz and a Q of 10^4 ; then at the frequency where the spectral density is 100 times below its maximum, that is 15 Hz from resonance, the error is still less than 0.3%.

IT IS EASY TO CHECK that $S_q(\omega)$ satisfies the equipartition theorem

$$\left\langle \frac{1}{2} k_0 q^2 \right\rangle = \frac{1}{2} k_0 \int_{-\infty}^{\infty} q(t)^2 dt = \frac{1}{2} \frac{k_0}{2\pi} \int_0^{\infty} S_q(\omega) d\omega = \frac{1}{2} k_B T, \quad (1.12)$$

where we used Plancherel's theorem, and the identity

$\int_0^{\infty} \frac{a}{(1-x^2)^2 + (ax)^2} dx = \frac{\pi}{2a}$ for Eq. 1.10, or $\int_{-\infty}^{\infty} \frac{a}{(x_0-x)^2 + a^2} dx = \pi$ for Eq. 1.11.

1.4 Heating

AS THE RESONATOR has a dissipation factor, it releases heat into the environment where the dissipation occurs. Our very cold materials and samples in the experiment can have very low heat capacities. Especially the spin or spin bath in the sample that couples to the resonator is very sensitive to heating. Therefore we should calculate if the heat production of the cantilever can raise the temperature significantly, or even dramatically. Let us consider two cases: a system that is driven with a sinusoidal force, and one that is excited with white noise around the resonance peak.

BY DEFINITION of the Q-factor,²³ the average power that is lost is given by

$$P_{\text{avg}} = \frac{1}{2} k_0 \langle q^2 \rangle \frac{\omega}{Q}, \quad (1.13)$$

For the sinusoidal force with frequency $\omega = \omega_d$ we have $\langle q^2 \rangle = (A(\omega_d))^2$, where A is defined in Eq. 1.8.

IF THE FORCE is coming from a thermal force,²⁴ the total energy in the resonator is $\frac{1}{2} k_B T_m$, where T_m stands for the mode temperature which characterizes the height of the thermal spectrum. The power induced into the sample where the dissipation occurs is

$$P_{\text{avg}} \approx \frac{1}{2} k_B T_m \frac{\omega_0}{Q}. \quad (1.14)$$

The sample also has a temperature, let's say T_s , and the fluctuations in the sample will induce movement in the cantilever until the system is in equilibrium ($T_m = T_s$). The rate at which this equilibrium process goes is $\frac{\omega_0}{2Q}$, and hence the net power going from the mode to the sample is $P_{m \rightarrow s} = k_B \frac{\omega_0}{2Q} (T_m - T_s)$.

However, it might be that the sample is cooled by the environment. Let us assume that this environment has a constant temperature T_h , the heat capacity of the sample is $C(T)$, and the rate at which the temperature energy transfers from sam-

²³ The Q-factor is generally defined as $2\pi \frac{\text{Total energy stored}}{\text{Energy lost per cycle}}$.

²⁴ Or we inject white noise, which looks like thermal noise.

ple to heat bath, and vice versa, is τ . When there is a steady flow of heat, i.e. T_m and T_h are fixed, then the temperature of the sample can be derived from the stationary condition $P_{m \rightarrow s} = P_{s \rightarrow h}$, which gives

$$\frac{\omega_0}{2Q} k_B (T_m - T_s) = \frac{1}{\tau} \int_{T_h}^{T_s} C(T') dT'. \quad (1.15)$$

To solve this equation we need to know $C(T')$. For $T_s \sim T_h$ we can use the fundamental theorem of calculus and find

$$T_s = \frac{\frac{\omega_0 \tau}{2Q} k_B T_m + C(T_h) T_h}{\frac{\omega_0 \tau}{2Q} k_B + C(T_h)}. \quad (1.16)$$

Usually the resonator will loose its energy in more than one area. For calculating the temperature of each different part of the sample, one should only use the contribution of that specific part to the dissipation, and thus replace $\frac{1}{Q} \rightarrow \Delta \frac{1}{Q_s}$, see Fig. 1.1.

FINALLY, we calculate the sample temperature for a specific situation. In this thesis, the sample is usually a semiclassical spin interacting with a magnetic tip on the resonator. The precise coupling and dissipation mechanism are further explained in Ch. 2. For a two-state spin, the heat capacity is $C(T) = k_B \left(\frac{\mu_s B_0}{k_B T} \right)^2 \cosh^{-2} \left(\frac{\mu_s B_0}{k_B T} \right)$, with μ_s the magnetic moment of the spin and B_0 the average (constant) magnetic field. If we in advance already use Eq. 2.11 for the dissipation factor $\Delta \frac{1}{Q}$, and assume $k_0 \langle q^2 \rangle \gg T_s \sim T_h$, we find

$$T_s \approx \left(1 + \frac{1}{2} \frac{|\mathbf{B}'_{\parallel \hat{B}_0}|^2}{B_0^2} \langle q^2 \rangle \right) T_h, \quad (1.17)$$

where $\mathbf{B}'_{\parallel \hat{B}_0}$ is the gradient of the magnetic field in the direction of the constant B_0 field. We assumed that the spin is connected to the heat bath with a relaxation time ($\tau = T_1$) longer than the resonator's period. The imposed assumptions show that the approximation is only valid when $\sqrt{\langle q^2 \rangle} \ll 2B_0 / |\mathbf{B}'_{\parallel \hat{B}_0}|$, which for typical values in this thesis leads to a maximal rms amplitude of 100 nm. Comparing this to the 0.05 – 0.5 nm which we would have when the can-

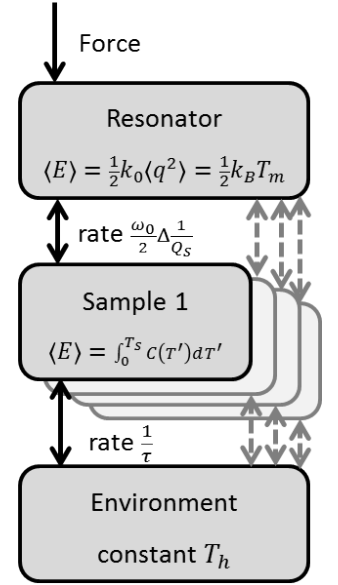


Figure 1.1: A thermodynamic schematic of the MRFM. The force working on the resonator causes a certain mode temperature. Sample 1, which is connected due to it's contribution to the Q-factor, is affected by the mode temperature. However, the sample is also (badly) connected to a heat bath. T_h is known, T_m can be measured, but what is T_s ?

tilever was thermalized to a heat bath of $0.01 - 1$ K, we see that it is not likely that the temperature of the spin bath is significantly changed due to the cantilever thermal motion. However, note that the mode temperature T_m is the sum over the squared movement of the resonator and can be significantly higher than T_h if the resonator is driven. In certain situations, such as during one of the OSCAR spin resonance protocols, the amplitude might be several 10s of nm,^{25,26} and the spin bath might heat up. When T_s becomes very different from T_h , we should recalculate T_s by solving Eq. 1.15. More on this subject is given in Ch. 2, where we take spin's resonance properties into account.

²⁵ Rugar et al. 2004

²⁶ Cardellino et al. 2014

1.5 Contents

APART FROM FORMULATING THE BASICS in this introduction chapter, we already touched on the main challenges that we need to overcome for creating an experiment that is able to measure gravitational collapse of the wave function. In our proposed experiment, where a macroscopic resonator is manipulated with the qubit, all we care about is a very good coupling between the resonator and the qubit, plus a very low dissipation of the mechanical resonator. We will show that the coupling can be very good. However, a central question, that needs to be answered in Ch. 2-4, can we also understand, control, or even avoid the dissipation? Only after we know that, it makes sense to find the optimal experiment as is explained in Ch. 5. On the other hand, numerous technical challenges that we have encountered and solved are summarized in Ch. 6. One of the most difficult parts of the MRFM experiments was, and still is, the three dimensional coarse approach at cryogenic temperatures. We tested the stability of the microscope that is used in Ch. 4 by measuring the stability of a tunneling current between a temporarily mounted Scanning Tunneling Microscope (STM) tip and a conductive sample.²⁷ As the approach of the tip to the sample had to be

²⁷ The STM-tip was mounted on the moving end of the coarse approach motor, while the Highly Oriented Pyrolytic Graphite (HOPG) sample was mounted on the base.

done without optical access, we monitored the approach by measuring the capacitance between tip and sample continuously. An analysis of this method turned into a relatively new technique which is useful for various Scanning Probe Microscopes. The article following from this spin-off side project is included as Ch. 7.