Magnetic resonance force microscopy for condensed matter
Wagenaar, J.J.T.

Author: Wagenaar, Jelmer J.T.
Title: Magnetic resonance force microscopy for condensed matter
Issue Date: 2017-07-05

3
A resonator magnetically coupled to a semi-classical spin

WE CALCULATE the change of the properties of a resonator, when coupled to a semiclassical spin through the magnetic field coming from a magnetic moment on the tip. Starting with the Lagrangian of the complete system, we provide an analytical expression for the linear response function for the motion of the resonator, thereby considering the influence of the resonator on the spin and vice versa. This analysis shows that the resonance frequency and effective dissipation factor can change significantly due to the relaxation times of the spin.

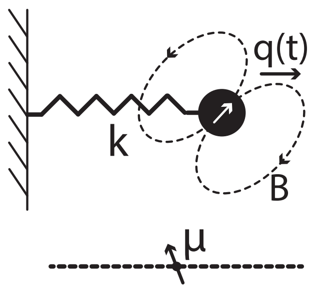

Figure 3.1: Schematic representation of spin $\boldsymbol{\mu}$ interacting with a mechanical resonator with spring constant k and displacement $q(t)$ of the magnet. The dashed line shows the position axis that is used in Fig.3.2.

THIS CHAPTER IS BASED ON J. M. DE VOOGD, J. J. T. WAGENAAR, AND T. H. OOSTERKAMP, "DISSIPATION AND RESONANCE FREQUENCY SHIFT OF A RESONATOR MAGNETICALLY COUPLED TO A SEMICLASSICAL SPIN", *Sci. Rep.*, VOL. 7, 42239, FEB. 2017

Resonators and spins are ubiquitous in physics, especially in quantum technology, where they can be considered as the basic building blocks, as they can collect, store and process energy and information[1]. The validity of this information is, however, of limited duration as these building blocks leak practically always to the environment, which on its own can be seen as a bath of resonators and spins[2]. If in particular we focus on the situation where a resonator is coupled to a certain spin, then the spin's interaction with the environment naturally causes, besides a shift of resonance frequency, an extra dissipation channel for the resonator. Despite this simple qualitative explanation and many experimental[3] and theoretical efforts[4], an applicable full picture that quantitatively describes the response of a resonator coupled to a spin and their environments is, to our knowledge, still lacking. Here we derive classically the linear response function of the non-conservative system consisting of a resonator and a semiclassical spin. We show that the quality factor and resonance frequency of the resonator can be significantly influenced due to the relaxation times of the spin.

[1] Nazarov and Blanter 2009; You and Nori 2005

[2] Caldeira and Leggett 1981; Prokof'ev and Stamp 2000

[3] Imboden and Mohanty 2009; Venkatesan et al. 2010; Bruno et al. 2015

[4] Caldeira and Leggett 1983; Sleator et al. 1987; Schlosshauer et al. 2008; Pappas et al. 2011

We start with a Lagrangian description, that includes the degrees of freedom of the resonator *and* the spin, to find the coupled equations of motion (EOMs) that describe the resonator displacement and the spin magnetic moment, finding that this magnetic moment depends on the path the resonator takes. This is fundamentally different from conventional magnetic force microscopy (MFM)[5], where one assumes a fixed polarization of the spins, as is indeed the case in magnetized samples. Even in magnetic resonance force microscopy (MRFM), which is usually focused on paramagnetic spins, it is generally assumed that the spin is not, or at least not significantly, influenced by the resonator[6]. We will show that this influence actually opens a dissipation channel and that the resonance frequency shift is more subtle than generally assumed.

[5] Rugar et al. 1990

[6] Rugar et al. 1992; Degen et al. 2009; Vinante et al. 2011a

Furthermore, we find in our analytical results that the interaction amplitude as function of temperature is a curve that for certain conditions shows an optimum, see Fig. 3.3, similar to the curves found in experiments where the tails have heuristically been fitted with power laws[7]. Parts of the analysis we present here have been used in Ch. 7 to explain the experimental results obtained by approaching a native oxide layer on silicon with an ultra-sensitive MRFM probe. The equations derived in this chapter were found to closely resemble the measured shift in resonance frequency and reduced quality factor as function of temperature and resonator - spin surface distance.

[7] Imboden and Mohanty 2009; Venkatesan et al. 2010

Finding an accurate description of the interaction of the building blocks of quantum devices with the environment can be seen as a widespread and major research area since not being able to understand, control and minimize the interaction is a major bottleneck in the field of quantum computing[8], detector fabrication in astronomy[9], MRFM and high resolution MRI[10] and the development of optomechanical hybrid quantum devices[11].

[8] Pappas et al. 2011; Bruno et al. 2015

[9] Day et al. 2003; Endo et al. 2013

[10] Poggio and Degen 2010; Kovacs et al. 2005

[11] Aspelmeyer et al. 2014; Lee et al. 2017

3.1 *Basic Principles*

The configuration of our theoretical analysis is given in Fig. 3.1a. A semiclassical spin, with magnetic moment $\boldsymbol{\mu}$, is located at laboratory position $\boldsymbol{r}_s$ and feels a magnetic field $\boldsymbol{B}(\boldsymbol{r}_s, t)$ that is produced by a magnet. The magnet is attached to a mechanical resonator that has spring constant k_0 and (effective) mass m_{eff}. The origin of the laboratory frame is chosen to be the equilibrium position of the magnet's center. The displacement of the magnet from this equilibrium position is denoted by $q(t)$. See Fig. 3.1. The Lagrangian for this system is given by

$$L = \frac{1}{2} m_{eff} \dot{q}^2 - \frac{1}{2} k_0 q^2 + \boldsymbol{\mu} \cdot \boldsymbol{B}(q) + I_S. \qquad (3.1)$$

I_S stands for an expression containing the internal spin degrees of freedom that needs to be included to derive the spin EOM[12].

[12] De Voogd et al. 2017

The resonator-spin system does not live in an isolated world. Therefore we include dissipation and decay to the environment into the EOMs. The first differential equation, derived with respect to the resonator displacement, includes the Raleigh dissipation $-\gamma \dot{q}$ of the resonator. This results in

$$m_{eff}\ddot{q} + \gamma\dot{q} + k_0 q - \boldsymbol{\mu} \cdot \frac{\partial}{\partial q}\boldsymbol{B} = F_{ext}(t), \qquad (3.2)$$

where the last term, $F_{ext}(t)$, is an external force that is exerted on the resonator.

Starting with the Lagrangian, which contains the degrees of freedom for the resonator *and* the spin, leads to the force interaction term $-\boldsymbol{\mu} \cdot \frac{\partial}{\partial q}\boldsymbol{B}$. This is the same as $-\boldsymbol{\mu} \cdot \nabla B_{\|q}$, because of the vanishing curl of the magnetic field. Here $\nabla \boldsymbol{B}_{\|q}$ is the gradient of the magnetic field component in the direction of the movement of the resonator. In MRFM $-\boldsymbol{\mu} \cdot \nabla B_{\|q}$ is often derived from calculating the force-field from the gradient of the potential energy $\nabla\left(\boldsymbol{\mu} \cdot \boldsymbol{B}\right)$, assuming that $\boldsymbol{\mu}$ does not depend on the position of the resonator[13]. However, as $\boldsymbol{\mu}$ follows the classical path, we will show by solving the spin EOM that $\boldsymbol{\mu}$ is influenced by the resonator and it is therefore a priori not at all obvious that $\frac{\partial}{\partial q}\boldsymbol{\mu} = 0$.

[13] Berman et al. 2006b

The other set of differential equations can be found by deriving the EOM with respect to the spin degrees of freedom. Since the spin interacts with the environment, we can expect an effectively decaying amplitude that is often described by T_1 and T_2; the time constants associated with the decay of the semiclassical magnetic moment longitudinal and perpendicular to the magnetic field, respectively[14]. If one assumes that the system consists of an ensemble of paramagnetic spins, instead of a single isolated spins, the average magnetic moment per spin decays to a certain equilibrium vector $\boldsymbol{\mu}_\infty$, according to the master equation[15]. However, if a single spin over time has on average the same behavior as the average of an ensemble at a certain moment, i.e. the spin satisfies ergodicity, then we can combine the ensemble's master equation and the single spin EOM to find a differential equation that describes the average behavior of the single semiclassical spin. This is the Bloch equation:

[14] Bloch 1946

[15] Slichter 1990

$$\dot{\boldsymbol{\mu}} = \gamma_s \boldsymbol{\mu} \times \boldsymbol{B} + T^{-1}\left(\boldsymbol{\mu}_\infty - \boldsymbol{\mu}\right). \qquad (3.3)$$

Here γ_s is the gyromagnetic ratio and $T^{-1} \equiv \frac{1}{T_2}\left(\mathbb{1} - \hat{\boldsymbol{B}}\hat{\boldsymbol{B}}^T\right) + \frac{1}{T_1}\hat{\boldsymbol{B}}\hat{\boldsymbol{B}}^T$, where the hat denotes the unit-vector in the direction of the specified vector and $\hat{\boldsymbol{B}}^T$ is the transposed unit vector.

The spin equilibrium magnetic moment $\boldsymbol{\mu}_\infty(t)$ is the vector to which the spin magnetic moment would decay to if given the time. As the resonator moves, the magnetic field changes, and so does $\boldsymbol{\mu}_\infty$. We will assume that the environment of the spin is a heat bath, connected to the spin by means of the relaxation times. However, does the spin's equivalent spin ensemble have a well defined temperature? As derived in the original paper of Bloembergen et al. (1948), the differential equation describing the population difference n for particles in a two level system is

$$\frac{dn}{dt} = -2Wn + \frac{n_0 - n}{T_1}, \tag{3.4}$$

where W is the rate that the particle changes energy level due to an applied field and n_0 is the population difference between the energy levels when the ensemble is in equilibrium, in others words when the ensemble has the temperature of the heat bath. $-2Wn$ is proportional to the incoming energy and $\frac{n_0 - n}{T_1}$ is the connection to the heat bath. This results in

$$n_\infty = \frac{n_0}{1 + 2WT_1}, \tag{3.5}$$

where n_∞ is the steady state solution. Thus when $2WT_1 \ll 1$, the spin ensemble, and hence our semiclassical spin, is connected well enough to the heat bath to assume that our spin has a well defined temperature. For spin-$\frac{1}{2}$ this condition yields[16]

[16] Bloembergen et al. 1948

$$\pi \gamma_s^2 \left|\boldsymbol{B}'\right|^2 q^2 T_1 g\left(\omega_0\right) \ll 1, \tag{3.6}$$

where $\boldsymbol{B}' = \frac{\partial}{\partial q}\boldsymbol{B}\Big|_{\boldsymbol{r}=\boldsymbol{r}_s}$ and $g\left(\omega\right)$ the spin's normalized absorption line that is usually described by a Lorentzian or Gaussian that peaks around the Larmor frequency at a value of 1. ω_0 is the resonance frequency of the cantilever at which the cantilever oscillated with amplitude q. This makes this condition hard to satisfy when the resonator has a resonance frequency around the Larmor frequency, and one should minimize the resonator's movement q.

[17] Slichter 1990

When this condition is not met, the spin saturates and the temperature increases or might be undefined[17]. However, for example in MRFM, mechanical resonators tend to have resonance frequencies much lower than the Larmor frequency and so it is much easier to satisfy this condition.
Assuming the condition is satisfied we can now derive $\boldsymbol{\mu}_\infty$ from the canonical ensemble and find for spin-$\frac{1}{2}$

$$\boldsymbol{\mu}_\infty = \mu_s \tanh\left(\beta\mu_s \left|\boldsymbol{B}\right|\right) \hat{\boldsymbol{B}}, \tag{3.7}$$

where $\beta \equiv \frac{1}{k_B T}$ is the inverse temperature and $\mu_s \equiv S\hbar\gamma_s$ is the magnitude of the non-averaged spin magnetic moment with spin number $S = \frac{1}{2}$. This result can easily be generalized for other spin numbers[18]. For simplicity we will stick to the formula for spin-$\frac{1}{2}$ particles here.

[18] De Voogd et al. 2017

3.2 *Susceptibility*

To find the resonance frequency and quality factor of the resonator, we will need to calculate the interaction term up to linear order in q. Higher order terms will give rise to nonlinear effects. Interaction terms with even powers in q are usually experimentally uninteresting since they will produce even multiples of the fundamental resonance frequency. These multiples are not measured or can easily be filtered. The higher order terms with uneven powers of q can, however, lead to disturbing nonlinear effects as in a Duffing oscillator[19]. One can lower the amplitude of q to suppress higher order terms and therefore the nonlinear effects, but in experiments this is usually limited by the signal-to-noise ratio.

[19] Kaajakari et al. 2004

The zeroth order term does not contribute to the dynamics of the system, however it does give rise to a constant deflection of the resonator. This can be solved by shifting the origin of the laboratory frame by the amount of the deflection; this causes, however, a (usually small) change of the coordinates of the spin.

To find the interaction term $-\boldsymbol{\mu} \cdot \frac{\partial \boldsymbol{B}}{\partial q}$ up to first order in q, we first need to solve Eq. 3.3 for $\boldsymbol{\mu}$ and find the constant and q-dependent parts. By substituting $q \to \lambda q$ we use

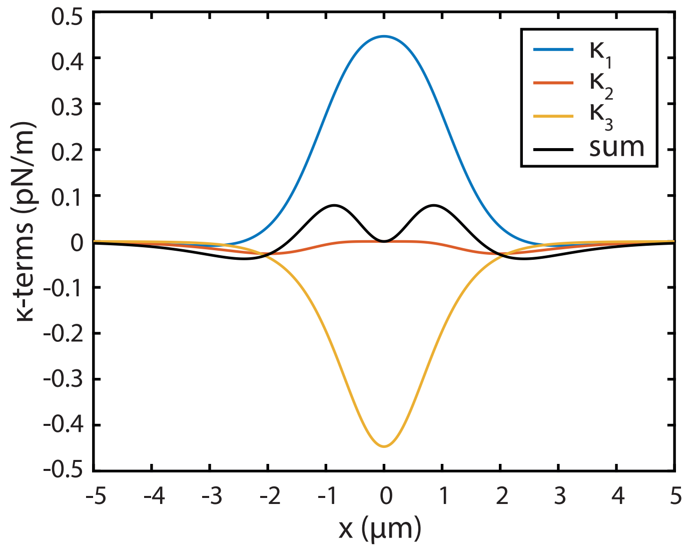

Figure 3.2: This graph shows the single spin contributions to the spring constant as function of a position axis parallel to the direction of resonator movement, as visualized by the dashed line in Fig. 3.1. In the simulation we attached a magnetic dipole (with magnetic moment of 19 pAm^2 in the direction of q) on a mechanical resonator. The resonator is connected, by means of the magnetic field, to an electron spin at a temperature of 300 mK. The distance between the center of the dipole and $x = 0$ is 2.5 μm. To demonstrate the spatial behavior of the κ-terms we avoided imaginary terms by setting $T_1 = 0$ in κ_2 and $T_2 = 0$ in κ_3. The black line shows the sum of these κ-terms.

perturbation theory to find that $-\boldsymbol{\mu} \cdot \frac{\partial \boldsymbol{B}}{\partial q}$ can be expressed as

$$-\boldsymbol{\mu} \cdot \frac{\partial \boldsymbol{B}}{\partial q} = \boldsymbol{\mu}_0 \cdot \boldsymbol{B}' + \lambda \left(\boldsymbol{\mu}_1 \cdot \boldsymbol{B}' - q\boldsymbol{\mu}_0 \cdot \boldsymbol{B}'' \right) + \mathcal{O}\left(\lambda^2\right), \tag{3.8}$$

where $\boldsymbol{B}' = \frac{\partial}{\partial q}\boldsymbol{B}\Big|_{\boldsymbol{r}=\boldsymbol{r}_s}$ was defined previously and $\boldsymbol{B}'' = \frac{\partial^2}{\partial q^2}\boldsymbol{B}\Big|_{\boldsymbol{r}=\boldsymbol{r}_s}$. Here $\boldsymbol{\mu}$ is perturbed into a q-independent part $\boldsymbol{\mu}_0$ and a linear term $\boldsymbol{\mu}_1$. The higher order terms $\mathcal{O}\left(\lambda^2\right)$ can be omitted, as well as the first term on the right hand side that only gives rise to the constant deflection.

At first we are mostly interested in solutions that do not decay over time and do not depend on initial conditions because then the linear response function can conveniently be given in the Fourier domain which makes it easy to compare with experiments. The Fourier Transform $\mathcal{F}\{\ \}$ of the linear response function, or simply susceptibility $\chi(\omega) \equiv \frac{\tilde{q}(\omega)}{\mathcal{F}\{F_{ext}\}}$, can be calculated from Eq. 3.2

$$\chi(\omega) = \frac{1}{k_0 - m_{eff}\omega^2 + i\gamma\omega + \kappa}, \tag{3.9}$$

where $\kappa = \kappa_1 + \kappa_2 + \kappa_3$, with $\kappa_1 \equiv -\boldsymbol{\mu}_0 \cdot \boldsymbol{B}''$ and $\kappa_2 + \kappa_3 \equiv \frac{\mathcal{F}\{\boldsymbol{\mu}_1 \cdot \boldsymbol{B}'\}}{\tilde{q}(\omega)}$. De Voogd et al. (2017) present the calculation of the κ-terms, which turn out to be:

$$\kappa_1 = -\mu_s \tanh\left(\beta\mu_s B_0\right) \left|\boldsymbol{B}''_{\|\hat{\boldsymbol{B}}_0}\right|,$$

$$\kappa_2\left(\omega\right) = -\frac{\mu_s}{B_0}\frac{\beta\mu_s B_0}{\cosh^2\left(\beta\mu_s B_0\right)} \left|\boldsymbol{B}'_{\|\hat{\boldsymbol{B}}_0}\right|^2 \frac{1}{1+i\omega T_1},$$

$$\kappa_3\left(\omega\right) = -\frac{\mu_s}{B_0}\tanh\left(\beta\mu_s B_0\right) \left|\boldsymbol{B}'_{\perp\hat{\boldsymbol{B}}_0}\right|^2 \times \left(1 - \frac{2\frac{T_2}{T_1} - \left(\omega T_2\right)^2 + i\omega T_2\left(1+2\frac{T_2}{T_1}\right)}{\left(1+i\omega T_2\right)^2 + \left(\omega_s T_2\right)^2}\right),$$

where $\boldsymbol{B}_0 \equiv \boldsymbol{B}\left(q=0\right)$ and the notation $\boldsymbol{v}_{\|\hat{\boldsymbol{B}}_0}$ and $\boldsymbol{v}_{\perp\hat{\boldsymbol{B}}_0}$ is used to indicate the part of $\boldsymbol{v}$ parallel and perpendicular to $\hat{\boldsymbol{B}}_0$ respectively for any vector $\boldsymbol{v}$. κ_2 and κ_3 are derived from $\boldsymbol{\mu}_{1_{\|\hat{\boldsymbol{B}}_0}}$ and $\boldsymbol{\mu}_{1_{\perp\hat{\boldsymbol{B}}_0}}$ respectively.

If we compare this result with the conventional approach that neglects the effect of the resonator on the spin, we see that in that approach we have only the term κ_1[20]. However, κ_1 is real and therefore it cannot describe the extra dissipation channel that has been seen in experiments[21]. The derivation which has been done here does include the linear effect of the resonator on the spin and vice versa. This produces two extra terms in the linear response function that are partly imaginary. Each of the κ-terms is shown separately in Fig. 3.2 as a function of the spin position. This position axis is indicated in Fig. 3.1 by the dashed line. Which effect these terms have in practice, where usually more than one spin is present, will be shown in the next section.

[20] Garner et al. 2004

[21] Vinante et al. 2011b; Wagenaar 2013a

3.3 *Spin Bath - Resonator Coupling*

We assume that all spins in the system act individually and do not influence each other, except through the relaxation times. We can then sum over the κ-terms for each spin to find the susceptibility of the resonator connected to a whole ensemble of spins, i.e. $\kappa = \sum_s \kappa_1(\boldsymbol{r}_s) + \kappa_2(\boldsymbol{r}_s) + \kappa_3(\boldsymbol{r}_s)$. Moreover, if the spins in the sample have an average nearest neighbor distance smaller than the typical spatial scale of the applied magnetic field, we can see the sample as a spin continuum and hence, instead of summing, integrate over the sample with spin density $\rho(\boldsymbol{r})$.

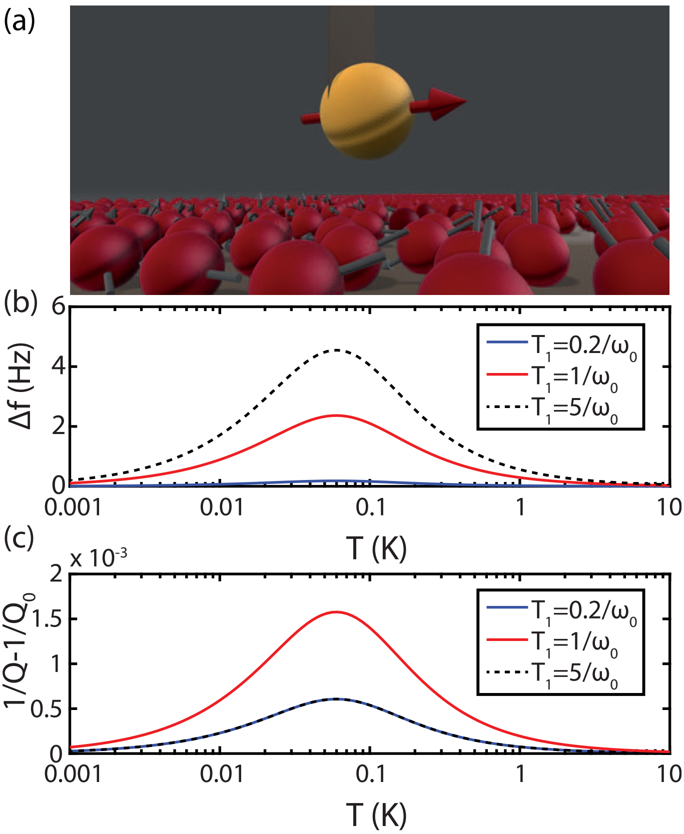

Figure 3.3: Calculated frequency shift and added dissipation of a mechanical resonator due to dangling bonds on a silicon surface, equivalent to the setup of Den Haan et al. (2015). **a)** Impression of a NdFeB magnet (with magnetic moment 19 pAm^2 in the direction of q) attached to an ultrasoft silicon cantilever with spring constant k = 70 µN/m, together leading to a natural frequency of $\frac{\omega_0}{2\pi}$ = 3 kHz. The surface of the sample has a native oxide containing 0.14 electron spins/nm^2 that are visualized by the red balls (not to scale). **b,c)** The resonance frequency shift and the damping of the cantilever as function of the temperature. The center of the magnet is positioned at a distance of 2.2 µm to the silicon sample. The results are shown for various T_1, showing a maximal opening of the additional dissipation channel for $T_1 = 1/\omega_0$. Note that in the lower graph that a the blue line and dashed black line lies on top of each other.

If we calculate the result for a volume with constant spin density, it can be found by partial integration of the volume in the direction of the movement of the resonator

$$\kappa(\omega) = \rho\beta\mu_s^2 C \frac{(\omega T_1)^2 + i\omega T_1}{1 + (\omega T_1)^2} + \text{boundary term} + \mathcal{O}\left(\frac{1}{(\omega_s^2 - \omega^2)\, T_2^2}\right), \quad (3.10)$$

with

$$C = \iiint_{\mathcal{V}} d^3\boldsymbol{r} \frac{\left|\boldsymbol{B}'_{||\boldsymbol{B}_0}\right|^2}{\cosh^2(\beta\mu_s B_0)}. \quad (3.11)$$

The boundary term vanishes when the volume boundaries in the $\boldsymbol{q}$-direction are large. The $\mathcal{O}\left(\frac{1}{(\omega_s^2-\omega^2)T_2^2}\right)$ can be neglected for resonance frequencies away from the Larmor frequency and for $T_2 \gg \frac{1}{\omega_s}$.
From κ we can calculate the frequency and Q-factor shifts as seen in experiments by Den Haan et al. (2015) (Ch. 7). For $Q_0 \equiv \frac{\sqrt{k_0 m_{eff}}}{\gamma} \gg \frac{1}{\sqrt{2}}$, the susceptibility has a maximum close to the natural frequency $\omega_0 \equiv \sqrt{\frac{k_0}{m_{eff}}}$. Then, as long as the influence of the spin leads only to a small correction of the susceptibility, i.e. $\kappa \ll k_0$, the relative frequency shift is given by

$$\frac{\Delta\omega}{\omega_0} \approx \frac{1}{2}\frac{\mathrm{Re}\,(\kappa(\omega_0))}{k_0}. \quad (3.12)$$

The imaginary part of κ causes the change in Q-factor. The new Q-factor is given by

$$\frac{1}{Q} \approx \frac{1}{Q_0} + \frac{\mathrm{Im}\,(\kappa(\omega_0))}{k_0}. \quad (3.13)$$

In Fig. 3.3 we show an example of an experiment with a magnet attached to an ultrasoft cantilever, which is positioned above a silicon sample. The native oxide contains electron spins that interact with the resonating magnet. The frequency shift and quality factor depend differently on T_1. In this simulation we have set T_2 to zero only after we checked that the $\mathcal{O}$ term in Eq. 3.10 can indeed be neglected: setting $T_2 = T_1$ gives an additional frequency shift of about 1 nHz and a five orders of magnitude lower shift in Q-factor compared to the results shown in Fig. 3.3c.

3.4 *Discussion and Conclusions*

We have calculated the linear response function of a mechanical resonator coupled to a spin. The linear response function of the resonator shows extra terms that result in a shift of the resonance frequency and a drop of the Q-factor of the resonator, compared to the bare resonator characteristics. In practice, this means that despite having resonance frequencies that are not even close to the Larmor frequency, one encounters dissipation of the resonator due to the inhomogeneous field it creates. Eventually this might not be a surprise since the resonator alters the heat capacity of the spin's equivalent spin ensemble. Although this is closely related to the magnetic loss enhancement in nonmagnetic glassy systems[22], we did not find any description in literature that provides a quantitative and detailed account of how this influences the linear response of the resonator, despite the many reported and unexplained results[23]. The results presented here have been experimentally verified[24] and have been used to calculate the frequency shift in a simple, yet powerful, saturation measurement protocol[25].

[22] Jug et al. 2016

[23] Imboden and Mohanty 2009; Venkatesan et al. 2010; Bruno et al. 2015

[24] Den Haan et al. 2015

[25] Wagenaar et al. 2016

We have chosen to do the calculations completely in the (semi)classical regime as we are especially interested in the expectation value of spin and resonator. Moreover this leads to an intuitive description and fairly simple calculations. The classical treatment has it limitations though: Berman et al. (2006a) have raised the point that in a quantum description, if the cantilever position is constantly measured, there is an influence on the spin because of the projections that are constantly occurring in the act of measuring. This might introduce random quantum jumps which, when they are not time averaged over timescales longer than T_1, are not taken into account in our description. Furthermore, when pulses are applied, for example in spin resonance techniques, a precise time evolution of the system is needed. Moreover, sending hard pulses might violate the condition for the temperature and linear response of the spin that we have encountered in Sec. 3.1. In this case one might move to a calculation involving the spin-operators. The theory presented here would still give a fair indication about the enhancement of dissipation, which is of importance in the field of hybrid quantum systems that are pushing the limit of macroscopic superpositions[26].

[26] Lee et al. 2017; Wezel and Oosterkamp 2012