Magnetic Resonance Force Microscopy at milliKelvin Temperatures

Wijts, G.H.C.J.

Author: Wijts, Geert
Title: Magnetic resonance force microscopy at milliKelvin temperatures
Issue Date: 2013-09-19

Chapter 2

Optimizing the force sensitivity in an MRFM experiment

In a Magnetic Resonance Force Microscopy (MRFM) experiment, we try to detect the force that is exerted on the magnetic tip of a soft cantilever by a small ensemble of spins. The force of a single spin is given by [17]

$$\vec{F}_m = \vec{\nabla}(\vec{\mu}_s \cdot \vec{B}_m) \tag{2.1}$$

Here, $\vec{\mu}_s$ is the magnetic moment of the spin and $\vec{B}_m$ is the magnetic field of the cantilever tip at the location of the spin. Table 2.1 lists the magnetic moments of an electron spin and a selection of nuclear spins, as well as the force they would exert on a typical MRFM cantilever. These forces are extremely small[1], ranging from a few zeptonewtons for nuclear spins to tens of attonewtons for electron spins.

In this chapter, we will discuss the relevant considerations for optimizing the signal-to-noise (SNR) ratio in an MRFM experiment, such that the sensitivity will be high enough to enable detection of such small forces. We will address why we need small magnetic tips, why we work with soft cantilevers with low damping factors, and under which conditions we expect the polarization of spins to be dominated by statistical polarization rather than by Boltzmann polarization.

2.1 The magnetic cantilever tip

It is evident from Eq. (2.1) that, in order to maximize the force the spin sample exerts on the magnetic cantilever tip, one should devise a tip that leads to a

[1] The electron spin force on the cantilever is smaller than the gravitational force a human being feels from a mosquito flying at a distance of 30 meters!

species	spin	γ [C kg^{-1}]	μ_s [μ_B]	F_m [10^{-18}N]
electron	1/2	$1.76 \cdot 10^{11}$	1.00	16.1
^{1}H	1/2	$2.68 \cdot 10^8$	$1.52 \cdot 10^{-3}$	0.0244
^{13}C	1/2	$6.73 \cdot 10^7$	$3.83 \cdot 10^{-4}$	0.00614
^{19}F	1/2	$2.52 \cdot 10^8$	$1.43 \cdot 10^{-3}$	0.0230
^{29}Si	1/2	$-5.32 \cdot 10^7$	$-3.02 \cdot 10^{-4}$	−0.00485
^{93}Nb	9/2	$6.57 \cdot 10^7$	$3.63 \cdot 10^{-3}$	0.0539

Table 2.1: Spin, gyromagnetic ratio γ, magnetic moment μ_s, and force F_m on a typical MRFM cantilever for electron spins and a selection of nuclear spins [11, 18]. The magnetic moment is given in units of the Bohr magneton $\mu_B = 9.274$ J/T. The force is calculated for a field gradient of $1.73 \cdot 10^6$ T/m, which is the maximum gradient at the surface of a NdFeB ferromagnetic sphere with a radius of 1.5 μm.

field gradient which is as high as possible at the location of the spins.

For a uniformely polarized spherical tip, the magnetic field as a function of position is given by [17]

$$\vec{B}_m(\vec{r}) = \frac{\mu_0}{4\pi r^3}\left(3\left(\vec{\mu}_m \cdot \hat{r}\right)\hat{r} - \vec{\mu}_m\right) \tag{2.2}$$

where μ_0 is the permeability of free space and $\vec{\mu}_m$ is the magnetic moment of the sphere. For a sphere with radius R, $\vec{\mu}_m = \frac{4}{3}\pi R^3 \vec{M}$, where $\vec{M}$ is the magnetization of the sphere.

The magnitude of the field gradient at the surface of the sphere scales as:

$$|\vec{\nabla}(B_m(r = R))| \propto \frac{M}{R} \tag{2.3}$$

Evidently, we should make the magnetic tip radius as small as possible in order to maximize the force it exerts on the spin sample, while using a tip material which keeps a high level of magnetization at the experimental conditions. MRFM experiments have been performed with micron-sized nickel spheres [19] glued to the cantilever, batch microfabricated cantilevers with 100 nm diameter nickel nanorod tips [20], 150 nm wide samarium cobalt tips [15], a 200 nm wide iron cobalt nanopillar deposited on a copper stripline with the spin sample on the cantilever tip [16], and, quite unconventionally, by sending a 67mA current through a 375 nm wide gold stripline, thus generating strong field gradients [21]. In this thesis, we describe work with micron-sized, ferromagnetic, NdFeB, spherical particles, which have a strong remanent field of 1.3 T [22,23]. A field gradient of 1.7 MT/m could be obtained by using a NdFeB sphere with a diameter of 3 μm.

2.2 Cantilever response to spin force

The motion $x(t)$ of the cantilever magnet along the easy axis of the cantilever is governed by the equation of motion:

$$m\ddot{x}(t) + c\dot{x}(t) + kx(t) = F_x(t) \tag{2.4}$$

where m is the effective mass of the cantilever-magnet assembly, concentrated at the cantilever tip, c is the damping factor, and k is the spring constant of the cantilever. For a force with a power spectral density $S_F(\omega)$ in N^2/Hz, the displacement power spectrum in m^2/Hz is given by

$$S_x(\omega) = \frac{S_F(\omega)/m^2}{(\Omega^2 - \omega^2)^2 + (\frac{\omega c}{m})^2} \tag{2.5}$$

where $\Omega = \sqrt{k/m}$ is the resonant frequency of the cantilever. In the limit of low damping factors $c/m << \Omega$, Eq. (2.5) can be approximated as a Lorentzian lineshape around the resonance [24]:

$$S_x(\omega) \approx \frac{S_F}{2kc} \frac{c/2m}{(\Omega - \omega)^2 + (c/2m)^2} \tag{2.6}$$

The Lorentzian lineshape has a Full Width at Half Maximum (FWHM) of $\Gamma = c/m$. The quality factor Q of the cantilever resonance is defined as $Q = \Omega/\Gamma = m\Omega/c$.

When we exert an sinusoidal force on the cantilever magnet at the resonant frequency Ω, the r.m.s. displacement of the magnet is $x_{rms} = \frac{F_{m,rms}}{c\Omega} = \frac{F_{m,rms}Q}{k}$. In order to maximize the cantilever displacement for a given force, the cantilever damping factor and resonant frequency have to be minimized, which is equivalent to choosing a cantilever that has a very low spring constant and a high quality factor. For a typical single crystalline silicon MRFM cantilever with a spring constant of 0.10 mN/m and a quality factor of $50 \cdot 10^3$, it follows from Tab. 2.1 that an electron spin and a proton spin can at most cause an r.m.s. displacement of 5.7 nm and 8.5 pm of our cantilever, respectively.

Although there are resonators with quality factors that are much larger than $50 \cdot 10^3$, such as thin silicon nitride membranes, which can have a fundamental mode quality factor as high as $Q_{\mathrm{SiN}} \approx 10^7$ [25], the stiffness of these resonators is usually so much higher ($k_{\mathrm{SiN}} \approx 30$ N/m), that their displacement for a given force is three orders of magnitude less than that of MRFM cantilevers.

2.3 Thermal force noise

Even in the absence of external forces on the cantilever, it exhibits random oscillations due to thermal force fluctuations. The power spectral density of this thermal force noise follows from the fluctuation-dissipation theorem that was put forward by Callen and Welton in 1951 [26]:

$$S_{F,th} = 4k_B T c = 4k_B T \frac{m\Omega}{Q} = 4k_B T \frac{\sqrt{km}}{Q} \tag{2.7}$$

The mean squared displacement of the cantilever magnet due to thermal fluctuations is

$$\left\langle x(t)^2 \right\rangle_t = \frac{1}{2\pi} \int_0^\infty S_x(\omega) d\omega = \frac{1}{2\pi} \int_0^\infty \frac{4k_B T c/m^2}{(\Omega^2 - \omega^2)^2 + (\frac{\omega c}{m})^2} d\omega = \frac{k_B T}{k} \tag{2.8}$$

This is in accordance with the equipartition theorem [27], which implies that the vibrational degree of freedom of the (classical) cantilever has an energy $\frac{1}{2}k\left\langle x^2\right\rangle = \frac{1}{2}k_BT$. For the MRFM cantilever mentioned above, which has an effective mass of about 0.28 ng, the amplitude spectral density of the force noise at room temperature is 41 aN/$\sqrt{\text{Hz}}$, resulting in an r.m.s. thermal vibration amplitude of 6.4 nm. This is already larger than the maximum possible displacement due to the oscillating force of a single electron spin. Hence, MRFM benefits from operation at low temperatures. At $T = 10$ mK, a temperature that is easily attainable with commercially available dilution refrigerators, the force noise can in theory be reduced to 0.24 aN/$\sqrt{\text{Hz}}$, which is equivalent to a thermal r.m.s. displacement of only 37 pm.

2.4 Spin polarization

For a nuclear species with spin I, the population density of the projection states $m = I, I-1, ..., -I$ onto the polarizing field is, in the thermodynamic limit, given by:

$$p_m = e^{m\hbar\gamma B_0/k_BT}\frac{\sinh\left(\hbar\gamma B_0/2k_BT\right)}{\sinh\left((I+1/2)\hbar\gamma B_0/k_BT\right)} \tag{2.9}$$

For a spin-$\frac{1}{2}$ then, the net polarization along B_0, also known as the Boltzmann polarization, is $P = \tanh\left(\frac{1}{2}\gamma\hbar B_0/k_BT\right)$. The room-temperature proton polarization in a state-of-the-art, 7T MRI scanner is only 24 ppm, so the number of spins needs to exceed the measurement sensitivity by a factor 42 thousand under those conditions. At cryogenic temperatures, the numbers become more friendly: at 10 mK, the proton polarization in a 7 T external field is 0.6, while at more moderate fields, such as the 0.87 T which is present at the surface of a NdFeB sphere, the Boltzmann polarization is still 0.09.

By striving for single-spin resolution in an MRFM experiment, requiring ever smaller spin ensembles for a detectable force, one moves away from the thermodynamic limit and enters a regime in which the incomplete cancellation of randomly polarized spins leads to a “statistical polarization”, which is described by $P_{\text{statistical}} \approx 1/\sqrt{N}$, where N is the number of spins [28, 29]. In Fig. 2.1 we have plotted the calculated number of spins for which statistical polarization is equal to Boltzmann polarization as a function of temperature, for various polarizing field strengths. If we, for example, consider an ensemble of hydrogen nuclei in a field of 0.1 T and at a temperature of 10 mK, we see that the statistical polarization becomes dominant for ensembles of less than ten thousand spins. For an electron spin under the same conditions, the Boltzmann polarization is dominant all the way down to the single spin ensemble.

We conclude that dilution refrigerator temperatures are advantageous for the polarization of small electron spin ensembles even in moderate field, but that for samples of less than 100 proton spins the temperature is basically irrelevant to the polarization, unless the polarizing field is very strong or if the temperature is in the sub-milliKelvin regime.

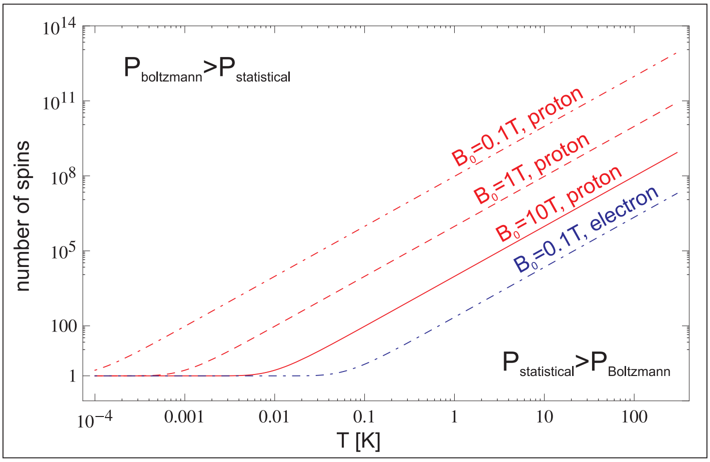

Figure 2.1: The number of spins for which the calculated statistical polarization is equal to the Boltzmann polarization as a function of temperature, for 3 values of the polarizing field strength B_0 and evaluated for proton spins (red lines) as well as electron spins (blue line). In the thermodynamic limit of very many spins, the Boltzmann polarization dominates, whereas the statistical polarization becomes relevant for small spin ensembles, even more so at higher temperatures and lower polarizing fields.

2.5 SNR in a force measurement

The Signal-to-Noise Ratio (SNR) is defined as the ratio between the signal power and the noise power in a given bandwidth. If the noise is completely random (spectrally "white"), the SNR can be increased by decreasing the measurement bandwidth down to the point where it is equal to the bandwidth of the signal. Additionally, by averaging over n independent measurements, the SNR can be improved by a factor $\sqrt{n}$.

In an MRFM measurement of the spin force, the bandwidth of the signal is determined by the coherence of the spin flips with respect to the oscillations of the cantilever. A correlation time τ_m is associated with naturally occurring spin fluctuations [30]. This sets a minimum for the measurement bandwidth.

The detection sensitivity of the cantilever displacement, usually determined by the electronic white noise in the detector, poses a maximum for a useful measurement bandwidth, which is the frequency range in which the cantilever thermal displacement noise exceeds the detection noise floor. In Fig. 2.2 we have plotted a simulation of the displacement power spectral density that illustrates this point. The blue curve represents the thermal motion of a cantilever with $k = 0.1$ mN/m and $Q = 50 \cdot 10^3$ at 10 mK. The red curve is the cantilever response to an alternating force with an r.m.s. amplitude of 1.6 aN and a correlation time of 1 s. The solid, black line is the detection noise floor at 10 pm/$\sqrt{\text{Hz}}$. Finally, the black, dashed line represents the measured displacement noise, adding all contributions. The noise floor is in this case chosen such that the detection bandwidth is equal to the signal bandwidth. If the noise floor were lower, the detection bandwidth would increase, allowing for the efficient measurement of force signals with lower correlation times. If the detection noise is increased, we would have to decrease the measurement bandwidth to maintain the SNR, but we would lose the sidebands of the signal.

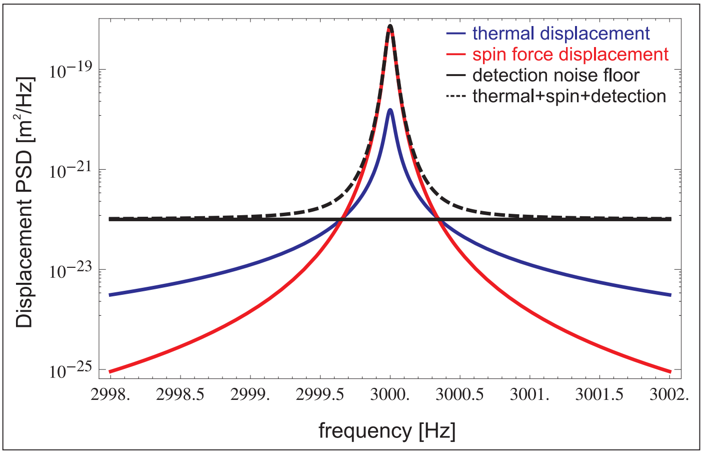

Figure 2.2: Simulation of the power spectral density measurement of the cantilever thermal vibrations including the thermal vibration noise (blue curve), a periodic spin force with a coherence time of $\tau_m = 1$ s at the cantilever resonant frequency (red curve), the detection noise floor (solid, black curve), and the cumulative signal resulting from all three sources (dashed, black curve). The simulation parameters are $k = 0.1$ mN/m, $c = 1 \cdot 10^{-13}$ Ns/m, $f_r = 3$ kHz, $F_{m,rms} = 1.6$ aN, $\tau_m = 1$ s, $T = 10$ mK, and $\sqrt{S_{det}} = 10$ pm/$\sqrt{\text{Hz}}$.

In Fig. 2.3 we have plotted the SNR of a force measurement of a single electron spin as a function of cantilever damping and temperature. Again, we assume a detection noise floor of 10 pm/$\sqrt{\text{Hz}}$ and a signal correlation time of $\tau_m = 1$ s. The curves represent the damping factor and temperature values for which the SNR= 1 in an infinitely narrow measurement bandwidth, for five different magnetic field gradients of the cantilever tip. As the tip gradient (and hence the signal power) increases, the restrictions on damping and temperature (which determine the noise power) become less stringent, allowing for higher values for both. The "knees" in the curves indicate at which temperature the cantilever thermal noise sinks below the detector noise. At lower damping factors, this point is pushed towards lower temperatures. The blue dot represents the experimental parameters that are attainable in our setup: a field gradient of ~ 1 MT/m, a damping factor of $1 \cdot 10^{-13}$ Ns/m and a temperature of 10 mK. We calculate from these numbers that we in principle can achieve an SNR of 157 in the force measurement of a single electron spin, with the requirement that $\tau_m = 1$ s.

For a single proton spin with an equally long correlation time, the single measurement SNR in our experiment is only $3.7 \cdot 10^{-4}$. By averaging 7.6 million times, an SNR of one can be reached, which, for a single measurement of 1 s, is equivalent to averaging for 90 days. For a statistically polarized ensemble

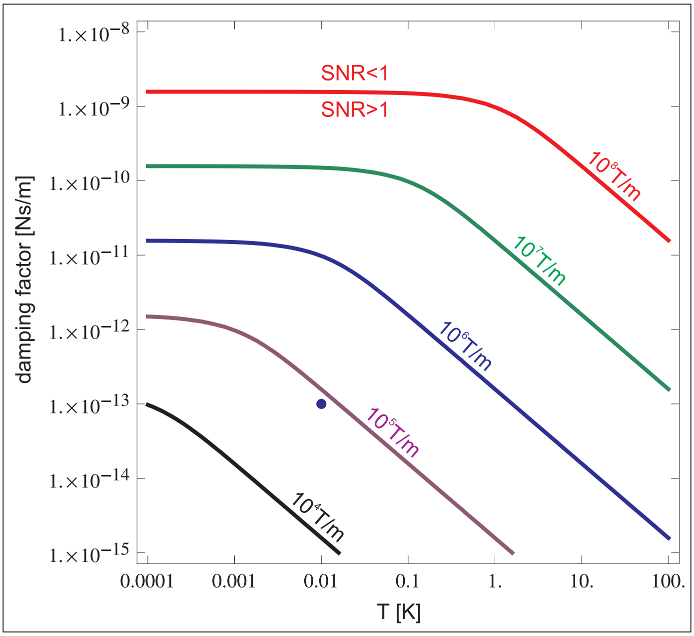

Figure 2.3: The curves in this graph represent the values of cantilever damping and temperature for which the SNR of the detection of a single electron spin in an MRFM measurement is equal to one, for several magnetic field gradients. The SNR is calculated for an infinitely small bandwidth. The plateaus in the curves correspond to values of cantilever damping and temperature where the detection noise is dominant over the thermal noise. We have used a displacement sensitivity of $\sqrt{S_{det}} = 10$ pm/$\sqrt{\text{Hz}}$ in this calculation. The blue dot corresponds to the parameters that are attainable in our experiment ($c = 1 \cdot 10^{-13}$ Ns/m, $T = 10$ mK, $|\vec{\nabla} B_m(\vec{r})| = 1.73 \cdot 10^6$ T/m), showing that an SNR>1 can be achieved.

of 100 protons, 1100 averages would suffice. An SNR larger than 1 can be achieved for an ensemble with a net polarization of 100 proton spins, or 10 thousand statistically polarized spins. In Fig. 2.1, we see that this is still in the regime in which statistical polarization dominates. For a biological sample, containing 40–80 protons per cubic nanometer, the detectable voxel size would be $(5.0\ \text{nm})^3$–$(6.3\ \text{nm})^3$.

No single improvement would make the detection of a single proton feasible, but if one could simultaneously improve one order of magnitude upon the magnet gradient (to 10 MT/m), the cantilever damping (to 10^{-14} Ns/m), and the temperature (to 1 mK), or alternatively one order of magnitude for the magnet gradient and two orders of magnitude for the cantilever damping, the quest for the holy grail of MRFM might be completed.

2.6 Conclusion

In this chapter, we have explored the optimization of the force sensitivity in an MRFM experiment. We have shown that the force signal can be maximized by devising a cantilever magnet that is as small as possible. The cantilever itself should be as soft as possible and have a high quality factor in order to be maximally displaced by an oscillating force. The experimental temperature should be ultralow in order to minimize the thermal force noise, which has as an additional advantage that electron spin samples or large ensembles of proton spins will be more strongly polarized. The force sensitivity in our experiment is already large enough to detect a single electron spin within a single measurement, provided that the signal correlation time is more than one second.