

# Theory of spin relaxation in magnetic resonance force microscopy

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We study relaxation of a spin in magnetic resonance force microscopy (MRFM) experiments. We evaluate the relaxation rate for the spin caused by high-frequency mechanical noise of the cantilever under the conditions of adiabatic spin inversion. We find quantitative agreement between the obtained relaxation time and the experimental results of B. C. Stipe, H. J. Mamin, C. S. Yannoni, T. D. Stowe, T. W. Kenny, and D. Rugar [Phys. Rev. Lett. **87**, 277602 (2001)]. Based on our analysis, we propose a method for improving the MRFM sensitivity by engineering cantilevers with reduced tip positional fluctuations. © 2003 American Institute of Physics. [DOI: 10.1063/1.1554769]

Magnetic resonance force microscopy (MRFM) has proven to be a powerful tool in studying magnetic properties of materials.<sup>1–4</sup> Unlike conventional magnetic resonance techniques, MRFM allows one to probe magnetization with a nanometer-scale spatial resolution, which is important for practical realization of spintronic<sup>5</sup> and quantum information processing devices.<sup>6</sup> Measurement of magnetizations produced by several hundreds of electronic magnetic moments has recently been reported.<sup>4</sup> Attempting to reach the single spin resolution, MRFM faces a number of experimental challenges. The observed reduction of the quality factors of the cantilever for small separations between the tip and the sample<sup>7,8</sup> and fast spin relaxation<sup>4</sup> pose a problem in resolving a single spin signal. In this letter, we analyze the latter effect. In particular, we study relaxation of a spin in the resonance slice<sup>4</sup> due to the thermal vibrations of the cantilever. We evaluate the spin relaxation rate and find a reasonable agreement with rates observed experimentally. We also propose that by shape engineering of the cantilever it is possible to filter the high-frequency noise thus reducing the spin relaxation.

In this work, we consider the experimental setup used in Ref. 4. One end of the cantilever is fixed, while the other end, to which a micron size magnetic particle is attached, is driven to oscillate in a direction normal to the surface of the sample; see Fig. 1. The sample is doped with spin 1/2 magnetic impurities. The smallest flexural-mode eigenfrequency of the cantilever will be denoted by  $\omega_0$  and the amplitude of the oscillations of the tip by  $z_0$ . In typical MRFM experiments,  $\omega_0$  is several kHz and  $z_0 \sim 0.1$ –30 nm. The motion of the magnetic tip of the cantilever produces an oscillating magnetic field  $\mathbf{B}_{\text{tip}}(\mathbf{r}, t)$  at the spatial position  $\mathbf{r}$  at time  $t$  (the origin is taken as the equilibrium position of the tip). There is an additional constant magnetic field  $\mathbf{B}_0$  applied normally to the sample, and a linearly polarized microwave magnetic field  $\mathbf{B}_1 \cos(\omega_{\text{rf}} t)$  in a direction perpendicular to  $\mathbf{B}_0$ .

The magnetic resonance condition is achieved as follows. Let us assume for simplicity that a single impurity spin is positioned at point  $\mathbf{r}$  immediately below the cantilever tip,

as shown in Fig. 1. Then, the magnitude of the field produced by the magnetic tip at the position of the spin is  $B_{\text{tip}}(\mathbf{r}, t) = B_{\text{tip}}^0(\mathbf{r}) + \nabla_z B_{\text{tip}}^0(\mathbf{r}) z(t)$ , where  $B_{\text{tip}}^0(\mathbf{r})$  is the field of the tip in equilibrium,  $z(t)$  is the displacement of the cantilever,  $|\mathbf{r}| \gg z(t)$ . Let the frequency  $\omega_{\text{rf}}$  of the microwave field be chosen such that  $g\mu_B |B_{\text{tip}}^0(\mathbf{r}) - B_0| = \hbar\omega_{\text{rf}}$ , where  $g$  is electronic  $g$  factor and  $\mu_B$  is Bohr magneton. Then, in the *rotating reference frame* that rotates with frequency  $\omega_{\text{rf}}$  around the normal to the surface,<sup>9</sup> the spin experiences an effective magnetic field (see the inset of Fig. 1)

$$\mathbf{B}_{\text{eff}} = \nabla_z B_{\text{tip}}^0(\mathbf{r}) z(t) + \mathbf{B}_1/2. \quad (1)$$

As a result, the spin precesses around this effective field, thus following the direction of  $\mathbf{B}_{\text{eff}}$ , as well as displacement of the cantilever, provided the latter varies with time adiabatically, i.e.,  $\omega_0 \ll \omega_{\text{eff}} = g\mu_B |\mathbf{B}_{\text{eff}}|/\hbar$ . Thereby, the spin direction follows the cantilever motion and the spin exerts an oscillating force on the tip of the cantilever with the amplitude,  $F = \mu_B \nabla_z B_{\text{tip}}^0$ . In Ref. 4, the shift of the frequency of the cantilever  $\Delta\omega_0$  due to the magnetization of the resonant spins has been measured. The signal that corresponds to roughly 100 fully polarized spins was observed to decay on a time scale of 100 ms, thus indicating that the induced magnetization of the resonant spins relaxes due to magnetic fluctuations or “noise” whose origin we will discuss.

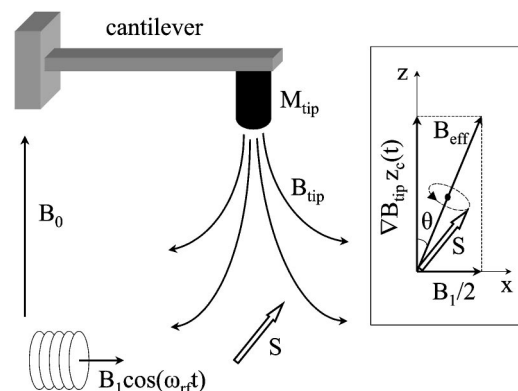


FIG. 1. Model setup for MRFM.

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In this letter, we show that the main source of the noise causing spin relaxation is likely to be related to the thermal vibration of the cantilever. The cantilever displacement can be written as  $z(t) = z_c(t) + \delta z(t)$ , where the first term,  $z_c(t) = z_0 \cos(\omega_0 t) + z_1$ , is due to the regular (driven) oscillation of the cantilever tip, while  $\delta z(t)$  is due to the thermal motion of the tip. Here,  $z_1$  defines the relative position of a spin within the resonant slice ( $|z_1| \leq z_0$ ). Substituting the aforementioned  $z(t)$  into Eq. (1) we obtain an effective Hamiltonian for a spin in the rotating frame

$$H = H_0(t) + n(t)s_z, \quad (2)$$

where  $H_0 = g\mu_B[\nabla_z B_{\text{tip}}^0 z_c(t)s_z + (B_1/2)s_x]$ ,  $s$  is the electronic spin, and  $n(t) = g\mu_B \nabla_z B_{\text{tip}}^0 \delta z(t)$ . In the following analysis, we set  $g = 2$  and the Planck constant  $\hbar = 1$ , unless stated otherwise. We assume that the noise  $n(t)$  is Gaussian with the correlation function  $K(t_1 - t_2) = \langle n(t_1)n(t_2) \rangle$  to be specified next. We derive the Bloch equations for the spin from the dynamics governed by the Hamiltonian (2). We introduce the Keldysh contour and define a real time partition function along the contour as

$$\mathcal{Z} = \mathcal{T}_c \exp\left(-i \int_{-\infty}^{\infty} H dt\right) \exp\left(-i \int_{-\infty}^{\infty} H dt\right), \quad (3)$$

where  $\mathcal{T}_c$  denotes ordering along the contour.<sup>10</sup> Thus, points on the forward branch ( $-\infty \rightarrow \infty$ ) are ordered with increasing times, while points on the return branch ( $\infty \rightarrow -\infty$ ) are ordered with decreasing times. The superscripts  $f$  and  $r$  on  $t$  will indicate to which contour  $t$  belongs. In Eq. (3), the time ordering operator  $\mathcal{T}_c$  sets operators on the return branch in front of the operators on the forward branch of the contour. The expectation value of an operator  $O(t)$  ( $O = s_x, s_y, s_z$ ) can be obtained by using the partition function  $\mathcal{Z}$  as  $\langle O(t) \rangle = \text{Tr}[\mathcal{T}_c O(t^f) \mathcal{Z}]$ , where the trace is taken over the states of the spin and over the distribution of the classical noise  $n(t)$ . Averaging the partition function  $\mathcal{Z}$  over the noise under the assumption that the noise is Gaussian, we obtain

$$\begin{aligned} \langle \mathcal{Z} \rangle_n &= \mathcal{T}_c e^{-i \int_{-\infty}^{\infty} H_0 dt} e^{-i \int_{-\infty}^{\infty} H_0 dt} \\ &\times \exp\left[-\frac{1}{2} \int_{-\infty}^{\infty} K(t_1 - t_2) s_z^a(t_2) s_z^a(t_1) dt_1 dt_2\right], \end{aligned} \quad (4)$$

where  $s_z^a(t) = s_z^f(t) - s_z^r(t)$ . The effective action for the spin defined by Eq. (4) is generally nonlocal. It can be significantly simplified by introducing the Bloch–Redfield approximation.<sup>9</sup> If the interaction of the system with the noise is weak, one can expect that on a scale of the noise correlation time,  $\tau_c$ , the evolution of the spin will be mostly determined by the unperturbed Hamiltonian  $H_0$ . That is, in the nonlocal part of the action one can replace  $s_z^a(t_2)$  by  $\exp[i \int_{t_1}^{t_2} H_0(\tau) d\tau] s_z^a(t_1) \exp[-i \int_{t_1}^{t_2} H_0(\tau) d\tau]$ . The latter operator can be evaluated assuming that  $H_0(\tau)$  varies adiabatically. By substituting the resulting expression into Eq. (4), the nonlocal part of the effective action in Eq. (4) can thus be approximated as

$$\begin{aligned} \Delta S = & -\frac{1}{2} \int \{ \cos \theta [\cos \theta s_z^a(t) + \sin \theta s_x^a(t)] S_n[0] \\ & + \sin \theta [\sin \theta s_z^a(t) - \cos \theta s_x^a(t)] S_n[\omega_{\text{eff}}(t)] \} dt, \end{aligned} \quad (5)$$

where  $S_n[\omega] = \int_{-\infty}^{\infty} K(t) \exp(i\omega t) dt$  is the power spectrum of the noise  $n(t)$ ,  $\omega_{\text{eff}}(t) = g\mu_B \{ [\nabla_z B_{\text{tip}}^0 z_c(t)]^2 + B_1^2/4 \}^{1/2}$ , and  $\sin \theta = g\mu_B B_1/2\omega_{\text{eff}}(t)$ . In the derivation of Eq. (5), we have assumed that  $\omega_{\text{eff}}/(d\omega_{\text{eff}}/dt) \gg \tau_c$ . The effective action in Eq. (5) is now local and allows for a straightforward derivation of equations of motion for the spin components  $s_{x,y,z}$ . Using  $\dot{s}_{x,y,z} = i[H_{\text{eff}}, s_{x,y,z}]$ , where  $H_{\text{eff}}$  is defined by Eqs. (4) and (5), and ordering the operators according to Eq. (3), we obtain a set of Bloch equations for the magnetization components

$$\dot{s}_x = -\omega_{\text{eff}}(t) \cos \theta(t) s_y + \beta(t) s_z - \alpha(t) s_x, \quad (6a)$$

$$\dot{s}_y = \omega_{\text{eff}}(t) \cos \theta(t) s_x - \omega_{\text{eff}}(t) \sin \theta(t) s_z - \alpha(t) s_y, \quad (6b)$$

$$\dot{s}_z = \omega_{\text{eff}}(t) \sin \theta(t) s_y. \quad (6c)$$

In Eq. (6), terms with coefficients  $\alpha$  and  $\beta$  describe relaxation of the magnetization due to noise,  $\alpha(t) = \{\cos^2 \theta S_n[0] + \sin^2 \theta S_n[\omega_{\text{eff}}(t)]\}/2$ , and  $\beta(t) = \sin 2\theta \{S_n[0] - S_n[\omega_{\text{eff}}(t)]\}/4$ . By solving the Bloch equations in the adiabatic limit, i.e., by assuming that the coefficients in Eq. (6) are slowly varying functions of  $t$ , we find that the component of the magnetization parallel to the effective field decays as

$$|s_{\text{eff}}(t)| \sim \exp\left\{-\frac{1}{2} \int_0^t \sin^2 \theta(\tau) S_n[\omega_{\text{eff}}(\tau)] d\tau\right\}. \quad (7)$$

Now we turn to the evaluation of the power spectrum of the magnetic noise produced by the cantilever tip. The magnetic noise is related to the mechanical noise of the tip as  $S_n[\omega] = (g\mu_B \nabla_z B_{\text{tip}}^0)^2 \int_{-\infty}^{\infty} dt \exp(i\omega t) \langle \delta z(0) \delta z(t) \rangle$ . It should be emphasized that we are interested in the mechanical noise of the cantilever at frequencies very high (of order  $\omega_{\text{eff}}$ ) compared to the lowest cantilever eigenfrequency  $\omega_0$ . As a consequence of the linearity of mechanical oscillator, the high-frequency modes of the cantilever will be essentially unaffected by the driving force at frequency  $\omega_0$ ; rather it is the thermal driving force that is important.

The energy of the vibrating cantilever can be expressed as  $E_c = \int_0^\ell dx [\rho(\partial_t z)^2 + EI(\partial_x^2 z)^2]/2$ , where  $z(x, t)$  is the transverse displacement of the cantilever at point  $x$  and time  $t$ ,  $\rho$  is the linear mass density of the cantilever,  $E$  and  $I$  are Young's modulus and moment of inertia of the cross section of the cantilever, respectively, and  $\ell$  is the length of the cantilever. The equation of motion for the free cantilever is  $\rho \partial_t^2 z = EI \partial_x^4 z$ .<sup>11</sup> The general solution to this equation can be written as  $z(x, t) = \sum_n z_n(t) \phi_n(x)$ , with  $\phi_n$ 's satisfying the eigenvalue equations for  $\omega_n$ :  $\rho \omega_n^2 \phi_n = EI \partial_x^4 \phi_n$ , supplemented by appropriate boundary conditions.<sup>11</sup> In the eigenfunction basis  $\phi_n$ , the energy of the cantilever can be expressed in terms of the amplitudes  $z_n$ :  $E_c = \sum_n [m \dot{z}_n^2 + m \omega_n^2 z_n^2]/2$ , where  $m = \rho \ell$  is the mass of the cantilever. Hence, the cantilever can be modeled as a collection of independent oscillators. In thermal equilibrium we then have that  $\langle \delta z_n^*(\omega) \delta z_n(\omega) \rangle = \delta_{n,n'} (\pi k_B T / m \omega_n^2) [\delta(\omega_n + \omega) + \delta(\omega_n - \omega)]$ , where  $k_B T$  is the temperature and the delta

functions  $\delta(\omega_n \pm \omega)$  have effective width of order of damping coefficient for the  $n$ th mode. The magnetic noise power spectrum can be expressed as

$$S_n[\omega] = (g\mu_B \nabla_z B_{\text{tip}}^0)^2 \sum_n \phi_n^2(\ell) \langle |\delta z_n(\omega)|^2 \rangle, \quad (8)$$

where the eigenfunctions  $\phi_n(x)$  are normalized,  $\int_0^\ell \phi_n^2(x) dx = \ell$ . The power spectrum given by Eq. (8) generally represents a set of distinct peaks. However, the effective frequency of a spin,  $\omega_{\text{eff}}(t)$ , sweeps over a large number of peaks. Moreover, the form of Eq. (7) indicates that it is the integral characteristics of the spectrum that are relevant for the determination of the spin relaxation rate. Therefore the sum in Eq. (8) can be replaced by the integral,  $\sum_n \rightarrow \int (\partial n / \partial \omega_n) d\omega_n$  and the finite width of the delta functions  $\delta(\omega_n \pm \omega)$  can be neglected. Then, taking into account that for  $n \gg 1$ ,  $\omega_n = (\pi n / \ell)^2 (EI / \rho)^{1/2}$ ,  $\phi_n^2(\ell) = 4$ , and  $\omega_0 = (3.52 / \ell^2) (EI / \rho)^{1/2}$ , see Ref. 11, Eq. (8) yields the average noise spectral density

$$S_n^{\text{ave}}[\omega] \approx 7.5 \frac{(g\mu_B \nabla_z B_{\text{tip}}^0)^2 k_B T}{m \omega_0^{1/2} \omega^{5/2}}. \quad (9)$$

Substitution of Eq. (9) into Eq. (7) gives the spin relaxation rate. For large times,  $t \gg \omega_0^{-1}$  and for  $\nabla_z B_{\text{tip}}^0 |z_0 \pm z_1| \gg B_1$ , the integral in Eq. (7) can be readily evaluated yielding  $|s_{\text{eff}}(t)| \sim \exp(-t/\tau_m)$ , where

$$\frac{1}{\tau_m} \approx \frac{3.4\mu_B \nabla_z B_{\text{tip}}^0}{\hbar} \left( \frac{k_B T}{k_s \sqrt{z_0^2 - z_1^2}} \right) \left( \frac{\omega_0}{\omega_1} \right)^{3/2}. \quad (10)$$

Here, we have introduced spring constant  $k_s = m\omega_0^2$  and  $\omega_1 = \mu_B B_1 / \hbar$ . Note, that spins located away from the center of the resonating region ( $z_1 \neq 0$ ) have faster relaxation rates. Therefore, measurement of an ensemble of spins is likely to yield a nonexponential MRFM signal decay.

We can now compare the relaxation time given by Eq. (10) with the experimental results of Ref. 4. For tip-sample separations of 800 nm the gradient of the magnetic field ( $\nabla_z B_{\text{tip}}^0$ ) was found to be roughly  $1 \times 10^5$  T/m. Assuming cantilever noise temperature  $T \sim 15$  K (Ref. 12), and taking other experimental parameters from Ref. 4, we obtain from Eq. (10) the maximum  $\tau_m \sim 70$  ms (for  $z_1 = 0$ ), which agrees well with the experimentally measured value.

In the mechanism discussed herein, the spin relaxation is caused by the thermomechanical noise of the high-frequency modes of the cantilever tip. This suggests a way to reduce the relaxation rate by engineering the shape of the cantilever to reduce this noise. The simplest approach that we analyze here is to place a massive particle ( $M$ ) on the tip. This will effectively filter out frequencies  $\omega_n$  for which  $n > m/M$ . For such high-frequency modes, it is easy to show that  $\phi_n^2(\ell) \approx 2\sqrt{\rho^3 EI} / (M^2 \omega_n)$ . For example, for a particle with  $M = m/10$  and the same experimental parameters, the relaxation time would increase to  $\tau \sim 5$  s. One cannot exclude, however, the possibility that other relaxation processes<sup>4</sup> may become the limiting factor in this regime.

In summary, we have found that high-frequency thermomechanical noise associated with high-frequency modes of the cantilever can induce significant spin relaxation in the MRFM, and is a likely reason for the coherent signal loss in the recent high-sensitivity experiments.<sup>4</sup> To reduce the influence of this noise, we have proposed cantilever shape engineering, which can lead to significant enhancement of MRFM sensitivity.

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