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Magnetic resonance force detection using a membrane resonator

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ABSTRACT

The availability of compact, low-cost magnetic resonance imaging instruments would further broaden the substantial impact of this technology. We report highly sensitive detection of magnetic resonance using low-stress silicon nitride (SiN_x) membranes. We use these membranes as low-loss, high-frequency mechanical oscillators and find they are able to mechanically detect spin-dependent forces with high sensitivity enabling ultrasensitive magnetic resonance detection. The high force detection sensitivity stems from their high mechanical quality factor $Q \sim 10^6$ [1,2] combined with the low mass of the resonator. We use this excellent mechanical force sensitivity to detect the electron spin magnetic resonance using a SiN_x membrane as a force detector. The demonstrated force sensitivity at 300 K is 4 fN/$\sqrt{\text{Hz}}$, indicating a potential low temperature (4 K) sensitivity of 25 aN/$\sqrt{\text{Hz}}$. Given their sensitivity, robust construction, large surface area and low cost, SiN_x membranes can potentially serve as the central component of a compact room-temperature ESR and NMR instrument having spatial resolution superior to conventional approaches.

1. Introduction

Magnetic resonance is a powerful tool that has had substantial impact on the fields of medicine, chemistry, and physics. Modern nuclear magnetic resonance (NMR) and magnetic resonance imaging (MRI) apparatuses utilize technology that has benefited from six decades of development, but their impact could be amplified in some settings by reducing their cost and physical size. Higher sensitivity can also enable enhanced spatial resolution in imaging applications.

Magnetic resonance force microscopy (MRFM) is based on mechanical detection of magnetic resonance signals. It has demonstrated imaging resolution far beyond that of the best inductive-based MRI [3–6], achieving nuclear-spin resolution better than ten nanometers [7,8]. The central component of MRFM is a mechanical resonator that is used to sensitively measure the force of interaction between the sample containing electron or nuclear spins, and a probe magnet with a strong field gradient. Depending on the experimental configuration, either the sample or the probe magnet is placed directly on the resonator [7,9] while the other component is in a fixed position in close proximity. The force of the probe-sample interaction is measured by detecting the displacement of the resonator via optical interferometry. Thermal force noise as low as 0.82 aN/$\sqrt{\text{Hz}}$ was achieved [10] using MRFM, culminating in an experiment that demonstrated single electron spin detection [9]. High sensitivity allows smaller volumes to be detected, hence enablihg high spatial resolution. At present such a high sensitivity has been achieved by using ultrasoft cantilevers with spring constants k as low as 110 μN/m [9]. While such cantilevers deliver exceptional force sensitivity, their use presents challenges: they are very fragile, sample preparation in the sample-on-cantilever geometry is difficult due to its small size, and such cantilevers are not commercially available.

Here we demonstrate that SiN_x membranes present a viable alternative to ultrasoft cantilevers as sensitive force detectors for MRFM applications. Such membranes exhibit a number of attractive properties including high sensitivity, robust mechanical properties, commercial availability, low cost, and a large surface area. The force noise of membranes can be as low as 8 aN/$\sqrt{\text{Hz}}$ [2,11,12], which is within an order of magnitude of the highest sensitivity demonstrated so far for an ultrasoft cantilever [10]. While less sensitive, the membrane is much less fragile because it is surrounded on all sides, unlike a cantilever supported only at one end. As a result, the membrane is much less susceptible to bending and twisting which otherwise can be detrimental for interferometric displacement detection. Samples can be quickly prepared on membranes utilizing similar sample preparation

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techniques as transmission electron microscopy (TEM), including application to the membrane by micropipette, or immersion in fluid on a glass slide. Finally, with their large surface area, membranes can accommodate a wider range of sample sizes and provide a larger target for interferometry than ultrasoft cantilevers.

One further advantage of membranes is their high natural frequency. The fundamental frequency of membranes can be in the MHz range, which enables resolving faster spin dynamics, and opens the door to new experiments. For example, since the Larmor frequency of nuclear spins in low field is also in the MHz range, there is the possibility of matching the membrane's mechanical resonance with the spins nuclear magnetic resonance frequency. This matching could overcome the current restriction to measuring only the *z*-component of the magnetization and allow force detection of its transverse component. This enticing possibility would provide MRFM access to the powerful array of imaging tools developed for pulsed NMR. Transverse detection of Larmor precession, known as direct-detection or "spin precession imaging" [13,14], could furthermore be accomplished without the necessity of an RF-generator. As the membrane oscillates, the spin sample on the membrane is physically displaced in the presence of a strong field gradient, which naturally generates the large oscillating magnetic field needed to excite the magnetic resonance signal. This innovation would aid in simplifying the MRFM apparatus. The apparatus could thus be reduced to three main components: a membrane, optical fiber-based displacement detection, and a magnetic particle on a translation stage as depicted in Fig. 1.

2. Experiment

2.1. Experimental details and setup

The MRFM experiment we report is performed by creating an oscillating force on a sample placed in the center of the membrane. The oscillating force is generated by modulating the sample magnetization using magnetic resonance, in the presence of a field gradient. The force drives the membrane at its natural frequency to an amplitude $A = FQ/k$, where F is the force, k is the spring constant, and Q is the quality factor of the membrane. We use the

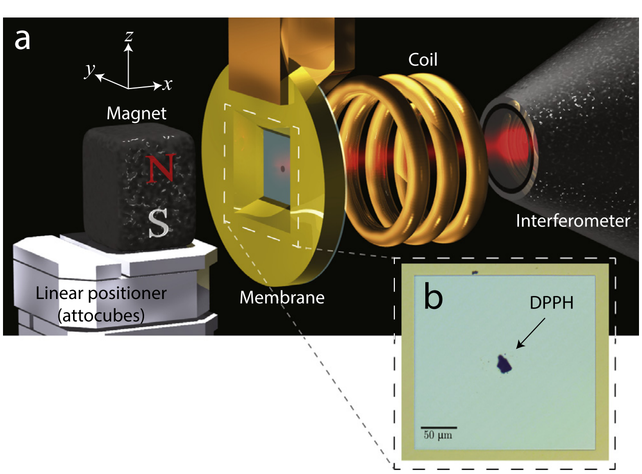

Fig. 1. Experimental schematic. (a) Schematic of the experimental setup. From the left, the interferometer laser passes through the copper coil and is incident on the membrane. A 20 μm particle of DPPH is placed in the center of the membrane. A permanent magnet produces both the polarizing field B_0 and a magnetic field gradient, which generates a force on the electron spins in the DPPH. (b) Photomicrograph of the 30 nm thick, 250 μm side-length silicon nitride membrane (inner turquoise square), surrounded by the silicon support. In the center of the membrane is the piece of DPPH is attached using a small amount of G1 epoxy. The bare membrane exhibits a frequency of 1.35 MHz, and the loaded membrane exhibits a frequency of 644 kHz.

cyclic-saturation resonance protocol [15,16] to measure a small particle of diphenyl picrahydrazyl (DPPH) on a membrane. DPPH is a well-known organic molecule that exhibits a electron paramagnetic resonance (EPR) signal.

The magnetic resonance signal is detected by measuring the displacement of the SiN_x membrane by means of a fiber optic interferometer, aligned as shown in Fig. 1. The interferometer uses 1550 nm laser light and is focused down to a 10 μm spot adjacent to the DPPH particle. The optical power incident on the membrane is about 80 μW. A 2.5 turn, 350 μm diameter copper resonance coil generates B_1, the RF field. The coil is centered on the membrane and is stub-tuned to a frequency $\omega_0/2\pi = 3.010$ GHz, setting the magnetic resonance condition $\omega_0/\gamma = 1070$ G, where γ is the electron gyromagnetic ratio. With an input power of 100 mW, the coil produces $B_1 = 4.7$ G. On the opposite side of the membrane, a two-axis piezoelectric stage [17] positions a rectangular NeFeB magnet, which provides both the polarizing magnetic field (B_0) and a field gradient of $G = 0.2$ G/μm. The instrument operates at a pressure of 10^{-6} torr.

Using the coordinate system in Fig. 1, the force on the electrons in the DPPH is given by $F = \mu_z \frac{dB_z}{dx}$, where $\mu_z = M_z V$, and V is the volume of the DPPH particle. The Bloch equations give the magnetization

$$M_z = M_0\left(1 - \frac{\gamma^2 B_1^2 \tau^2}{1 + (\gamma B_0 - \omega)^2 \tau^2 + \gamma^2 B_1^2 \tau^2}\right), \tag{1}$$

where $M_0 = \frac{\chi_0 B_0}{\mu_0}$ is the thermal equilibrium magnetization, B_0 is the magnetic field in the *z*-direction generated by the permanent magnet, $\chi_0 = 2.5 \times 10^{-5}$ is the susceptibility of DPPH, μ_0 is the permeability of vacuum, $\tau = 62$ ns is the spin relaxation time, and $\frac{\gamma}{2\pi} = 28$ GHz/T is the electron gyromagnetic ratio. To create the oscillating force, we induce an oscillating moment at the membrane frequency by modulating the frequency ω of B_1 such that $\omega(t) = \omega_0 + \Omega \sin(2\pi f_c t)$, where f_c is the membrane frequency. This results in a time-varying magnetization $M_z(t)$ whose Fourier component M_1 at the membrane frequency is given by $M_1 = \Omega \frac{\partial M_z}{\partial \omega}$ [15]. The derivative leads to a bipolar line shape of the force as a function of B_0.

The magnetic resonance signal is measured by varying the position of the permanent magnet and hence the magnitude of the applied magnetic field experienced by the sample on the membrane. The region of the magnet's field where the resonance condition is satisfied ($B = 1070$ G) is referred to as the "resonant slice," the thickness of which is $\Delta B/G \sim 10$ μm, where $\Delta B \sim 2$ G is the linewidth of DPPH. Due to the gradient, each spin in the resonant slice experiences a slightly different field, so the force as a function of magnet position is an integral as described in reference [16]; see the appendix.

2.2. Force noise

The central component of the MRFM apparatus is a sensitive mechanical oscillator which is employed as a force detector whose force sensitivity is limited by thermal force noise S_F given by

$$S_F^{1/2} = \left(\frac{2kk_B T}{\pi Q f_c}\right)^{1/2}, \tag{2}$$

where k is the spring constant, k_b is the Boltzmann constant, T is the temperature, Q is the quality factor, and f_c is the natural frequency. It is illuminating to cast Eq. (2) in terms of intrinsic membrane parameters such as thickness, side length, tensile stress, and density; t, L, σ, and ρ, respectively. The frequency is given by $f_0 = \sqrt{\frac{\sigma}{2\rho L^2}}$, and the spring constant is given by $k = \frac{\pi^2 \sigma t}{2}$ [18], yielding