Scanning Probe Microscopy in an Ultra-Low Vibration Closed-Cycle Cryostat: Skyrmion Lattice Detection and Tuning Fork Implementation

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Introduction

Many-body phenomena such as recently imaged currents in topological insulators [1] or nanoscale spin textures in chiral magnets, so-called magnetic skyrmions [2], arise at cryogenic temperatures constraining scanning probe microscopy (SPM) to be low-temperature compatible and to operate in liquid helium based cryostats [3, 4]. However, the high costs of helium and its scarcity have become a limiting factor, recently propelling the development of closed-cycle cryostats [5, 6]. These are independent of a continuous supply of liquid helium but present a challenge to the implementation of SPM owing to their noisy mechanics. Tuning fork-based scanning gate microscopy in a pulse-tube-cooled dilution refrigerator has been recently reported [7], opening the way to SPM in dry cryostats. This pioneering achievement required heavy intervention on the cryo-cooling system, sacrificing thermal contact and requiring long pre-cooling times. In this article we report on the development of an ultra-low vibration closed-cycle cryostat that allows top-loading of a variety of scanning probe microscopes with rapid turnovers. Because the microscopes do not require spring-mounting to further reduce vibrations, high-resolution confocal light microscopy imaging can be combined with scanning force microscopy. The ultra-low amplitude of vertical vibrations measured in our system enabled the AFM resolution of 0.39 nm high atomic steps in a $SrTiO_3$ sample. This low level of vibration also allowed detection of the skyrmion-lattice phase in a chiral magnet. Furthermore, it opened the way to the implementation for the first time of tuning fork-based shear-force microscopy in a closed-cycle cryostat. Scanning tunneling microscopy measurements recently reported by Leiden Cryogenics BV complete the picture of SPM measurements in a closed-cycle cryostat [8]. This article describes the cryo-cooled AFM and gives some example images.

Materials and Methods

Cryo-cooling system. Our dry cryo-cooling system (Figure 1) relies on conductive cooling and is based on pulse tube technology [6]. Here, a cryogenic fluid at high pressure is pulsed longitudinally through the different stages of the tube at a frequency of 1.4 Hz, reaching temperatures of approximately 3 K. The cyclic expansion and compression of the gas in the adiabatic chamber where the fluid is stored produces a stratification of temperature along the tube, so that one end is warmer than the other. The Cryomech PT-410 2-stage pulse tube cryo-cooler is driven by a compressor, which produces 1 W of cooling power at 4.2 K. A rotary valve regulates the pressure of the gas through the pulse tube. A 9 T superconducting magnet completes the system and is cooled by the pulse tube through thermal contact. Initial cooling of the whole system requires 12 hours; cooling down of the microscope insert from T=300 K to base temperature, namely 4 K, is normally achieved within 1 hour, depending on the mass of the microscope. One hour is also required to warm the system up. The sample space is immediately accessible after warming up thanks to the top-loading architecture. The system configuration enables measurement turnovers every 3 hours: sample exchange times of typically a few minutes are included in this estimate. The SPM experiments and the characterization of the vibrations in the system take place in the 50 mm diameter sample space, consisting of air-tight, removable, thin-walled

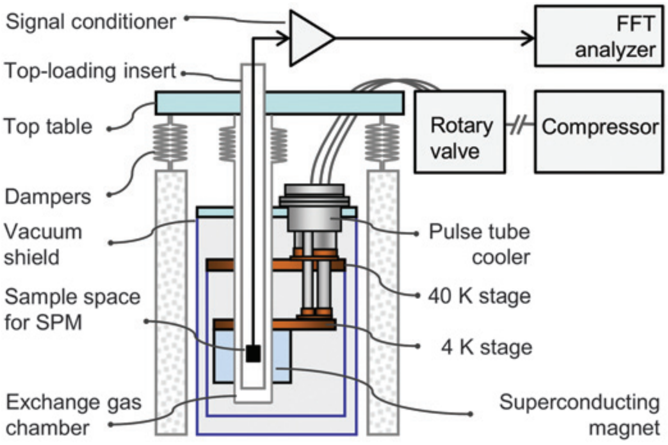

Figure 1: Closed-cycle cryo-cooling system based on pulse tube technology. The system consists of three main parts: a pulse tube, a rotary valve, and a compressor. A 9T superconducting magnet, fixed to the low-temperature stage of the pulse tube, surrounds coaxially the sample space, and it is cooled by thermal contact. A vibration detector in its vacuum insert is top-loaded into the system to characterize vibrations in the sample space.

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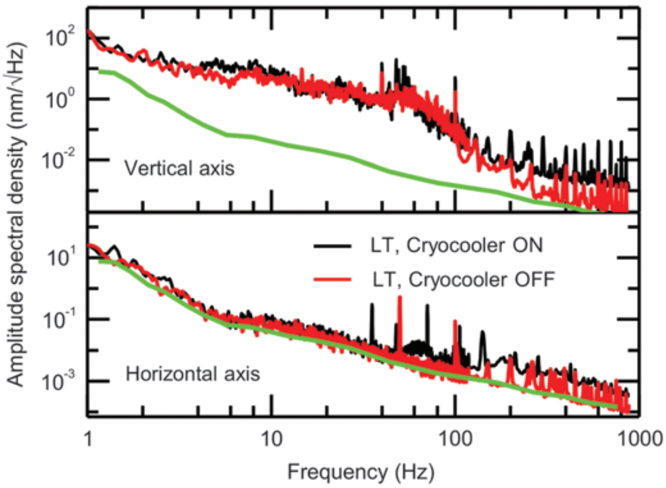

Figure 2: Vibration amplitude measured in the cryostat at T = 4 K with the cryo-cooler ON (black line) and OFF (red line). The top panel accounts for the vibration amplitude along the vertical axis; the bottom panel shows the amplitude of the vibrations along the horizontal axis. The green line represents the background level of vibrations measured independently in an isolated environment where we detect the lowest level of vibrations in our laboratory.

stainless steel tubing, hosting the microscope insert. Figure 1 shows mechanical decoupling between the pulse tube and the sample space in such a way that the vibrations from the pulse tube cryo-cooler do not influence vibration-sensitive experiments, while still ensuring a good thermal contact for sufficient cooling power.

Measurements of vibrations. Vibration amplitudes in the system were measured using an absolute vibration detector [9]. Figure 2 shows the spectral vibrational noise density along the vertical and the horizontal axis, measured in the sample space at T = 4 K. The response of the vibration detector at low temperature was found to be consistent with its performance at room temperature. The vibration detector is inserted in the vacuum tube at the location of the scanning probe microscope to simulate experimental conditions (Figure 1). Figure 2 shows measurements for each direction of displacement corresponding to the cryo-cooling system on (black line) and off (red line). The green line represents the background level of vibrations measured independently in an isolated environment where we detect the lowest level of vibrations in our laboratory. The data are shown on a logarithmic scale to visualize the amplitude spectral density. Figure 3 shows the absolute vibration amplitude between 1 Hz and 800 Hz, revealing the technical spurious noise. The rms vibration amplitude between 1 Hz and 800 Hz yields a peak value above background noise of 6 nm along the vertical axis and 3.5 nm along the horizontal axis at the pulse tube frequency f = 1.4 Hz. The peak at f = 50 Hz and its harmonics are parasitic electromagnetic pick-up of the vibration detector and do not correspond to vibrations. This is verified by comparison between the measurements with the cryo-cooling system on and off. The measured values of absolute vibration amplitude along the vertical axis in the nm range are low enough to enable SPM measurements in contact mode atomic force microscopy (AFM) and magnetic force microscopy.

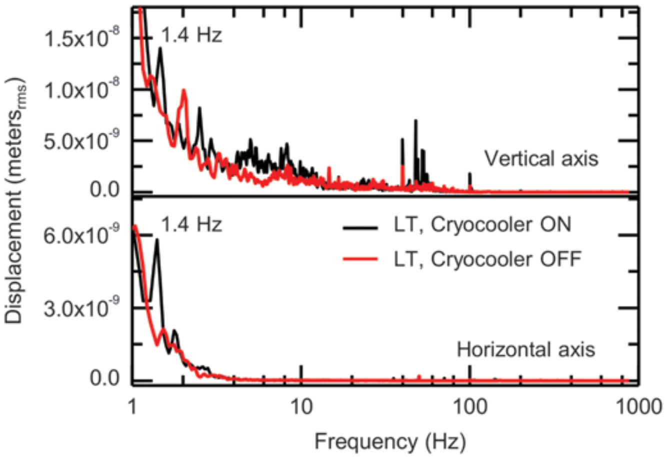

Figure 3: Measurements of displacement determined by the vertical (top) and horizontal (bottom) residual vibrations in the sample space inside the closed-cycle cryostat. Amplitude is measured at base temperature with the cryo-cooler ON (black line) and the cryo-cooler OFF (red line).

Results

Contact-mode AFM measurements. For contact mode AFM an etched silicon cantilever with a silicon tip combined with optical deflection detection was used to measure local interactions such as van der Waals or Coulomb forces. We used fiber-based interferometric sensing [10] to detect the deflection: a fiber was mounted in the head of the microscope to detect the deflection of the cantilever induced by the tip-sample interaction. A measurement of the amplitude of the relative displacement between the AFM tip and the sample was performed to evaluate the impact of the absolute vibrations of the system on the performance of the scanning probe experiments. A schematic representation of our AFM head with interferometric read-out is shown in Figure 4a. The *z*-noise was measured while keeping the tip in contact with the sample surface. Vibration data describing the relative displacement between the tip and the sample for the fully enabled system were then recorded over time. Noise statistics of the tip deflection were acquired over 10,000 points. The sampling time was set to 5 ms, corresponding to a measurement bandwidth of 200 Hz. Figure 4b shows the noise histogram. The result is a normal Gaussian distribution of the noise with a standard deviation σ = 65 pm rms; this value, measured with the feedback loop enabled, is comparable to the *z*-noise amplitude obtained in liquid systems [11]. When the feedback is turned off, the noise amplitude increases by approximately a factor of 4. To demonstrate the low vibration noise, images were taken at T = 3.2 K on terraces of height matching the lattice parameter (a = 0.39 nm) of a strontium titanate ($SrTiO_3$) commercial wafer, shallow polished at 0.1° then annealed to obtain terrace-and-step rearrangement at the surface. The image in Figure 5a shows a contact mode scan revealing the terrace-and-step morphology of the sample surface, with steps measuring 0.4 nm. The image was acquired in 21 ms sampling time, with a bandwidth of 47.6 Hz. These measurements are fully consistent with those reported of a similar commercial sample of $SrTiO_3$ measured at room temperature, showing 0.4 nm lattice steps without the influence of a cryo-cooling system [12].

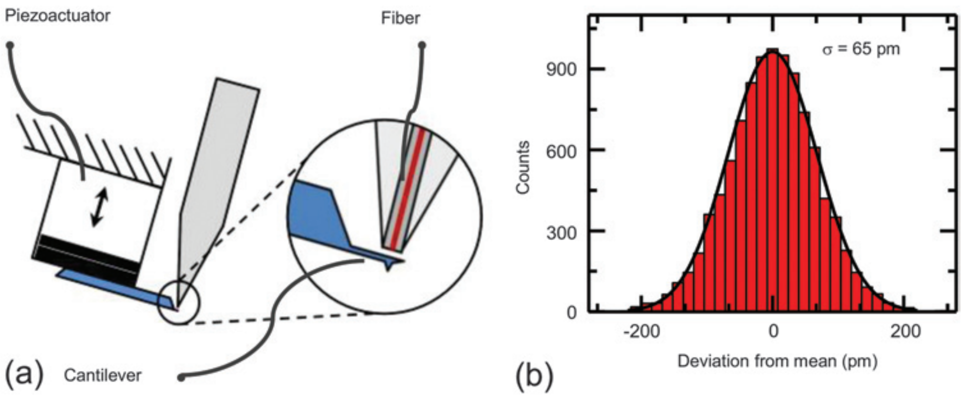

Figure 4: (a) Interferometric head for AFM measurements. (b) Contact mode noise scan histogram of the *z*-height values measured with a bandwidth of 200 Hz at 3.2 K. Measured amplitude of the relative displacement was 65 pm.

Magnetic force microscopy measurements. Exciting research in low-temperature solid-state physics is currently carried out by means of magnetic force and magnetic resonance force microscopy, including the imaging of currents in topological insulators [1], nanoscale spin configurations (magnetic skyrmions) in chiral magnets [2], scanning diamond magnetometry using nitrogen-vacancy (NV) centers in diamonds as solid-state sensors of magnetic field [13–15], and scanning gate microscopy [7, 16]. These techniques require sensitivity to extremely small magnetic interactions with nanometer spatial resolution. Their recent successful implementation required liquid bath cryostats. Given the scarcity of liquid helium, the implementation of scanning magnetic force microscopy inside dry cryostats represents the way forward. The low level of vibrations achieved in our dry cryostat allowed a variety of magnetic force microscopy measurements.

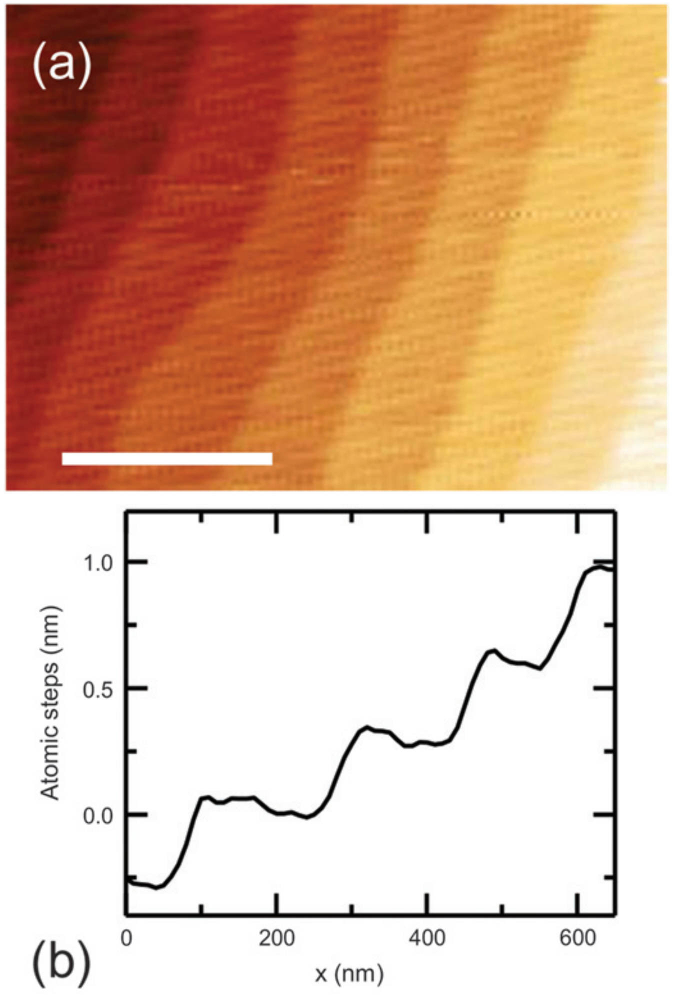

Figure 5: (a) Contact-mode AFM image of terraces on $SrTiO_3$ (200 scan lines) at 3.2 K. Bar = 400 nm. Step height is 0.39 nm, corresponding to the lattice parameter of the crystal. Frame time for the acquisition was 1,680 s at a scan rate of 500 nm/s. (b) Single-line profile showing the height of the atomically flat terraces on $SrTiO_3$.

In magnetic force microscopy, tips with magnetic coatings, typically NiCr or cobalt (Co), are employed that are sensitive to variations of magnetic field. The tip is oscillated at its resonance using a dither piezo; as it is scanned across the sample, the strength of the magnetic interaction between the tip and the magnetic fields near the surface determines a shift in the oscillation frequency. Frequency shifts map the magnetic structure of the sample. Figure 4a shows a diagram of the cantilever-based force microscope used in this work, specific for applications at low temperature and high magnetic field. Magnetic force microscopy is especially sensitive to the sample-tip separation, and vertical vibrations can affect image resolution by impairing the force sensitivity of the probe. We measured skyrmion lattices in our dry cryostat with an unprecedented signal-to-noise ratio of 20:1.

Skyrmion-lattice and helimagnetic phase in single-crystal $Fe_{0.5}Co_{0.5}Si$. Magnetic skyrmions are nanoscale spin whirls. Hexagonal lattices of these whirls are observed in certain chiral magnets without inversion symmetry in a phase pocket in finite magnetic field [17, 18, 19]. Perhaps most excitingly, electric currents allow these magnetic textures to move already at ultra-low current densities [20, 21]. Combined with their size in the nanometer range and their stability arising from the topologically non-trivial winding, skyrmions promise great potential for applications in information technology [22]. In addition to the bulk chiral magnets, skyrmions are recently also studied in thin films and monolayers, where they are typically driven by the surface and may be readily studied by means of SPM [23, 24]. For this report, however, we investigated the surface of a polished single crystal of $Fe_{0.5}Co_{0.5}Si$ in our dry cryostat [2]. We observed (Figure 6a) helimagnetic structures at T=3.2 K in zero magnetic field and a skyrmion-lattice texture (Figure 6b) at T = 3.4 K in an externally applied field B = 15 mT. The measurements of the helimagnetic phase in $Fe_{0.5}Co_{0.5}Si$ and the imaging of the skyrmion-lattice phase transition of the single crystal were carried out using a sharp tip (tip apex radius ~10 nm) with a magnetic coating (Nanosensors, SSS-MFMR). The magnetic tip was kept at a constant height of 20–30 nm over the sample surface, with a phase-locked loop activated to monitor the cantilever resonance frequency. The sample was heated to 60 K, and the magnetic field was

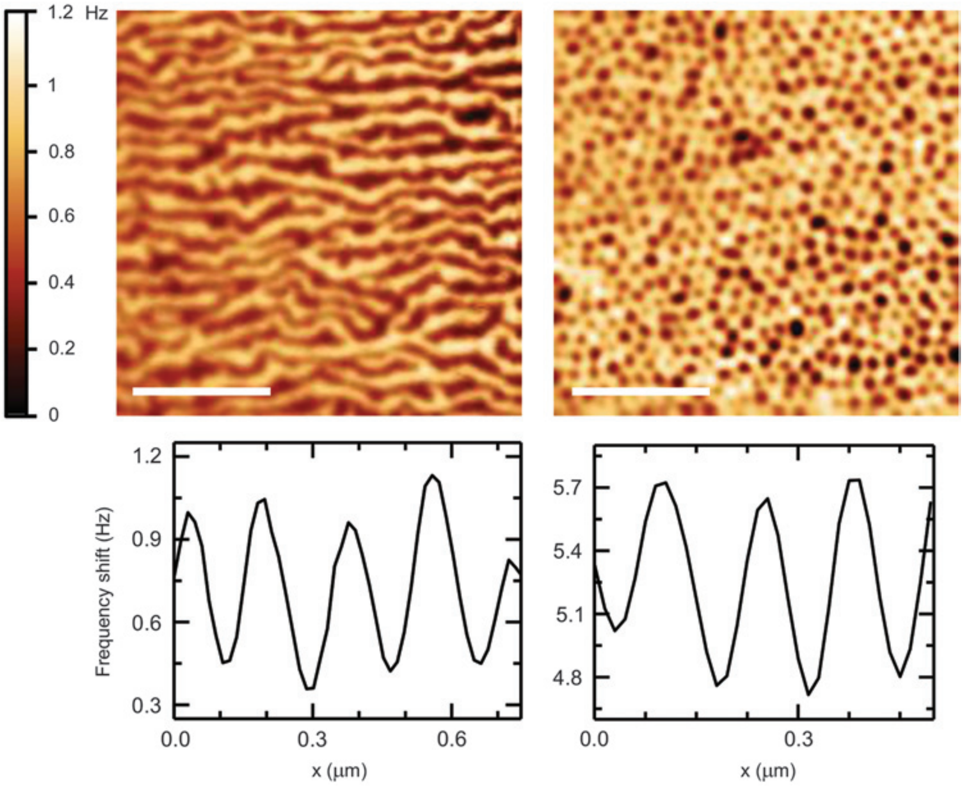

Figure 6: MFM images of the polished surface of a bulk sample of $Fe_{0.5}Co_{0.5}Si$. Images consist of 200 scan lines acquired using a sharp commercial cantilever (tip apex radius ~10 nm, SSS-MFMR from Nanosensors) with magnetic coating. (a) Helimagnetic phase of the sample at T = 3.2 K after zero-field cooling (B = 0). (b) Metastable skyrmion-lattice phase measured at T = 3.4 K in an external magnetic field B = 15 mT after field cooling. Bars correspond to a length of 1 μm. Line cuts highlight the frequency shift.

increased to 15 mT. The sample was subsequently field-cooled to base temperature again. With the persistent switch heater of the superconducting magnet enabled, the temperature stabilized at 3.4 K at the sample position. Figure 7 shows an image sequence of the coalescence of the skyrmion phase into helimagnetic phase. The magnetic field was decreased from B = 15 mT, where the metastable skyrmion lattice was observed, to B = −30 mT, where the textures coalesce into the helimagnetic structure [2].

Tuning Fork Scanning Force Microscopy measurements. Magnetic force microscopy imaging may be carried out with sensors exhibiting higher force sensitivity, such as tuning forks [7]. We show shear-force tuning fork measurements performed in our dry cryostat, which demonstrate the potential for further improvement, in particular toward the use of NV-centers in diamond-based scanning probe magnetometry [13].

The implementation of quartz tuning forks as self-sensing probes in scanning force microscopy is especially beneficial because of their small size, the absence of thermal effects induced by the optical detection, and the avoidance of unnecessary exposure of sensitive samples to light at cryogenic temperatures. To the best of our knowledge this is the first time that tuning-fork shear-force microscopy measurements have been successfully reported in a dry cryostat. A tungsten tip was etched in house to reach a tip apex radius of approximately 80 nm and glued to one prong of the tuning fork [25] as shown in Figure 8a. The viscoelastic interaction of the oscillating fork takes place through shear forces between the sample and the tip. Akiyama probes [26] are quartz resonators where a sharp silicon microcantilever is placed at the ends of the prongs, preserving the symmetry of the system. Typical values for the Akiyama spring constant are 5 N/m. The low noise amplitude measured between the tuning fork and the sample, with a bandwidth of 200 Hz and the feedback loop enabled, shows a normal Gaussian distribution with a standard deviation σ = 2 nm (Figure 8b). We demonstrate the achievement of tuning fork scanning force microscopy measurements of 20 ± 2 nm high SiO_2 patterns on Si with both Akiyama probes and in shear-force mode in our closed-cycle cryostat at T = 4 K (Figure 9a and 9b, respectively). The resolution is 200 lines per scan, acquired at 500 nm/s. Scanning in shear-force mode in a liquid-based cryostat, we measured noise amplitude better than 0.1 nm rms and, as discussed earlier, the contact mode noise measured in our dry cryostat was 65 pm.

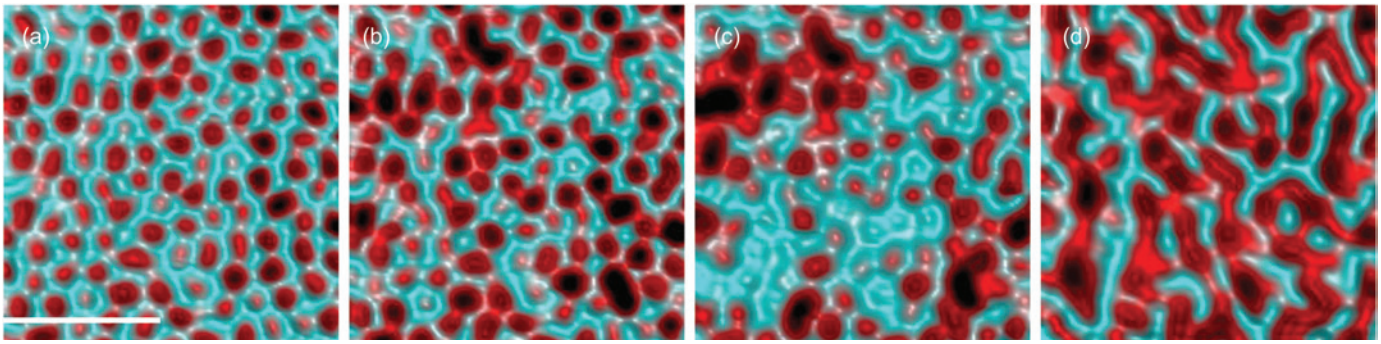

Figure 7: Coalescence of skyrmion-lattice phase into helimagnetic phase in $Fe_{0.5}Co_{0.5}Si$ with decreasing magnetic field B = 15 mT to −30 mT. Bar = 500 nm. Acquisition details as stated in **Figure 6**.

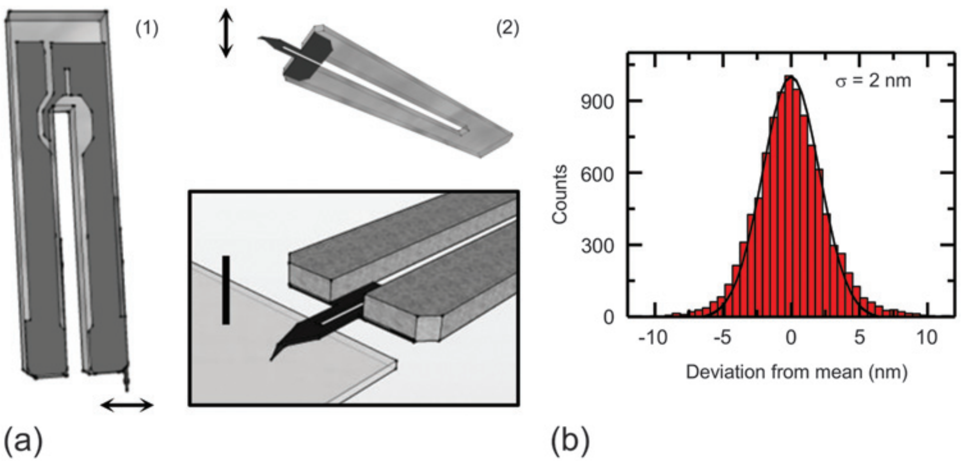

Figure 8: (a) Quartz-crystal tuning fork with tungsten tip (1) and Akiyama probe (2). Bar = 0.2 mm. (b) Shear-force mode noise scan; we measured 2 nm rms noise (bandwidth 200 Hz).

Discussion

The ultra-low vibration environment in our dry cryostat enables immediate application of SPM techniques. Contact-mode AFM measurements inside the dry cryostat resolved atomic steps of 0.39 nm height in a $SrTiO_3$ sample, owing to a relative tip-sample vibration amplitude of less than 65 pm. This level of noise allowed measurement of the skyrmion-lattice phase in $Fe_{0.5}Co_{0.5}Si$ by magnetic force microscopy. This had only been achieved quite recently in state-of-the-art liquid cryostat systems [2]. Furthermore, quartz-crystal tuning fork shear-force microscopy has been achieved for the first time to the best of our knowledge in a dry cryostat.

We studied the absolute vibrational noise in the sample space using a true inertial technique, measuring residual vibrations in the low nm range. We have also shown that these small displacements at very low frequencies do not influence the operation of the microscope and still enable measurements with high sensitivity. Our microscope is top-loaded in the cryostat and does not require being spring-suspended for further vibration isolation. This stiff architecture of the microscope head is of particular advantage when combining SPM with high-resolution confocal light microscopy where free-space optics is incompatible with spring mounting. The system as described allows a turnover of up to three experimental sessions per nine-hour working day. Beyond this work we hope to enable the implementation of further high-resolution imaging methods in dry cryostats and novel instances of magnetic force microscopy, such as scanning diamond magnetometry [13].

Conclusion

A closed-cycle dry cryostat capable of maintaining temperatures as low as 4 K was used as a platform for magnetic force microscopy measurements. This instrument provided images of nanoscale spin textures in chiral magnets such as magnetic skyrmions. Tuning-fork shear-force microscopy measurements in a dry cryostat are shown here for the first time. The stiff architecture of the system allows for the combination of SPM with light optical imaging, by achieving vibrations as low as 65 pm.

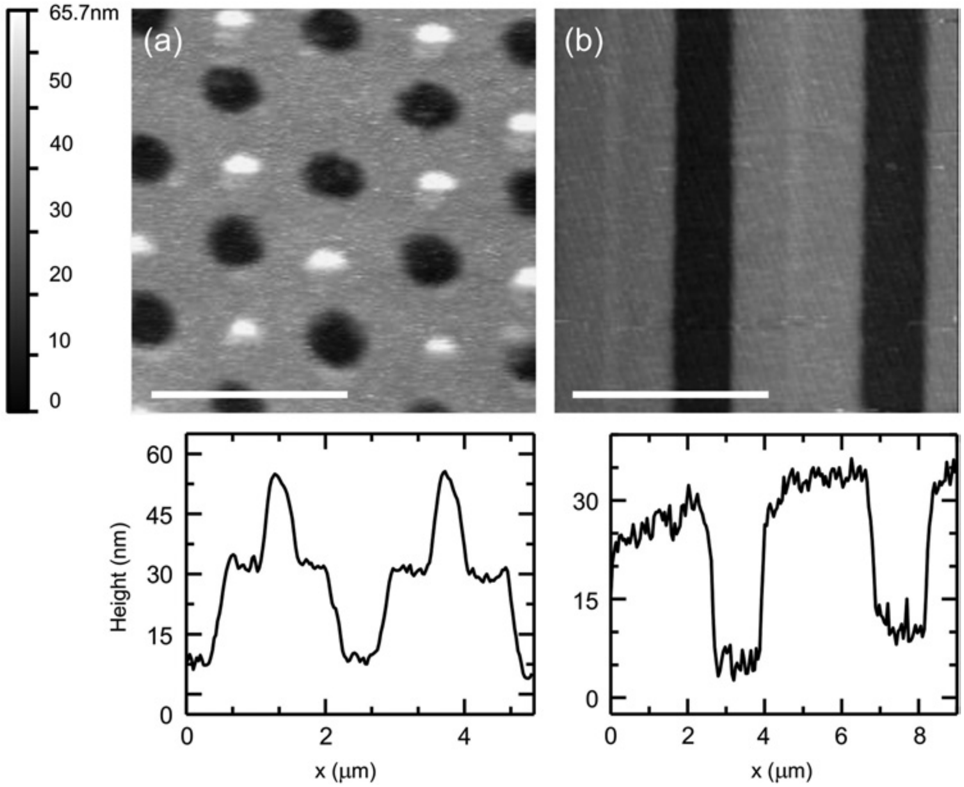

Figure 9: Quartz-crystal tuning fork AFM scans of SiO_2 20 ± 2 nm high patterns on Si, performed in our dry cryostat at T = 4 K. 200 scan lines at 500 nm/s. (a) 5 × 5 µm^2 tapping mode scan with Akiyama probe (test grating pitch 2 µm). B = 2 µm. (b) 9 × 9 µm^2 shear-force mode scan with quartz-tuning-fork-tungsten-tip configuration (test grating pitch 4 µm). Bar in (d) = 4 µm. Cross sections relative to each scan are shown.

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References

[1] K Nowack et al., *Nature Materials* 12 (2013) 787–91.
[2] P Milde et al., *Science* 340 (2013) 1076–80.
[3] D Rugar et al., *Nature* 430 (2004) 329–32.

[4] HJ Mamin et al., *Nature Nanotech* 2 (2007) 301–06.
[5] R Radebaugh, *J Phys–Condens Mat* 21 (2009) 164219–28.
[6] A Chijioke and J Lawall, *Cryogenics* 50 (2010) 266–70.
[7] M Pelliccione et al., *Rev Sci Instrum* 84 (2013) 033703–14.
[8] AMJ den Haan et al., *Rev Sci Instrum* 85 (2014) 035112–18.
[9] JA Stafford, Seismic vibration detector. US Patent 2651769 A, filed Aug 1, 1952, issued Sept 8, 1953.
[10] D Rugar et al., *Appl Phys Lett* 55 (1989) 2588–90.
[11] attocube systems AG, *Product Catalogue 2013 & 2014*. attocube, München, Germany, 2013.
[12] M Huijben et al., *Adv Mater* 21 (2009) 1665–77.
[13] G Balasubramanian et al., *Nature* 455 (2008) 648–51.
[14] S Hong et al., *MRS Bulletin* 38 (2013) 155–61.
[15] HJ Mamin et al., *Science* 339 (2013) 557–60.
[16] AGF Garcia et al., *Phys Rev B* 87 (2013) 085446–11.
[17] S Mühlbauer et al., *Science* 323 (2009) 915–19.
[18] W Münzer et al., *Phys Rev B* 81 (2010) 041203(R).
[19] XZ Yu et al., *Nature* 465 (2010) 901–04.
[20] F Jonietz et al., *Science* 330 (2010) 1648–51.
[21] T Schulz et al., *Nature Physics* 8 (2012) 301–04.
[22] A Fert et al., *Nature Nanotech* 8 (2013) 152–56.
[23] S Heinze et al., *Nature Physics* 7 (2011) 713–18.
[24] N Romming et al., *Science* 341 (2013) 636–39.
[25] K Karrai and RD Grober, *Appl Phys Lett* 66 (1995) 1842–44.
[26] http://www.akiyamaprobe.com.

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