Nanoscale Imaging Gone Dry

Sensitive scanning force microscopy in closed-cycle cryostats allows studying quantum phenomena.

Florian Otto and Christoph Bödefeld

Scanning probe microscopy (SPM) at cryogenic temperatures and in high magnetic fields allows studying many quantum phenomena at the nanoscale. Due to the ongoing helium crisis, closed-cycle cryostats are becoming more and more popular. However, almost all available dry coolers suffer from severe vibrations, which prohibits sensitive measurement techniques. In this article, we present state-of-the-art SPM measurements performed in a new type of dry magnet system with extremely low vibrations. This cryostat system provides a unique solution for sensitive techniques that require a calm, yet cold environment.

Many quantum phenomena studied in physics as well as materials science today require variable or cryogenic temperatures, often in conjunction with high magnetic fields. In order to reach low temperatures and to cool superconducting solenoids, which provide high magnetic fields, helium is the only available cooling agent. For more than a hundred years now, scientists have made use of liquefied helium and successfully employed its low boiling point of 4.2 K to cool their samples. Even lower temperatures can be achieved by pumping on liquid helium to reduce its vapor pressure. However, liquid helium has never been cheap, and in recent years, prices have gone up considerably. At the same time, supplies have become increasingly unreliable even in high-tech locations such as some of Europe's capitals. This insecurity has started to threaten the scientific progress of many scientists, and hence the move towards the so-called "dry" technologies is a natural evolution.

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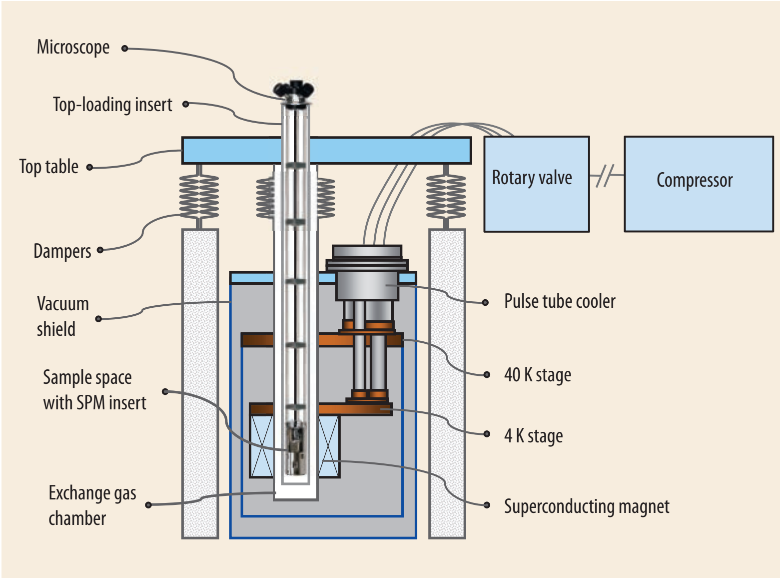

Fig. 1 The closed-cycle cryocooling system is based on pulse tube technology and consists of three main parts: a pulse tube, a rotary valve, and a compressor. A 9 T superconducting magnet, fixed to the lower temperature stage of the pulse tube, surrounds the sample space coaxially and is cooled by thermal contact.

Such cryostats still have no other choice than to cool with helium, but they keep a fixed amount of the precious gas contained in a closed-cycle, which maintains low temperatures by a cyclic compression and expansion of the working gas. Hence, the running costs are kept at a fraction of those compared to liquid helium based coolers. The invention of the Gifford-McMahon and pulse-tube coolers seems to have solved most problems at first sight. However, one very significant problem has remained with almost all commercially available setups: The cold heads generate relatively strong vibrations on the order of many microns, which is prohibitive for most sensitive measurement techniques – in particular scanning probe microscopy, which extends down to (sub-)atomic resolution routinely.

In this article, we present a new cooling system, which by design minimizes the vibrations dramatically, and hence enables even delicate measurements without further modifications. In particular, the microscopes do not require spring-mounting or active dampening inside to further reduce vibrations. This even enables the combination of scanning force microscopy with high-resolution confocal imaging and spectroscopy using free optical beams, as well as combinations with magnetic fields. We show some recent examples of state-of-the-art applications of scanning probe microscopy based research performed in an exceptionally quiet dry cryostat.

The Cryostat

The design of attocube's dry cryocooling system (**Fig. 1**) is based on a pulse tube cooler, with two separate cooling stages at $T = 40$ K (top stage) and $T = 4$ K (lower stage). The 2-stage pulse tube cryocooler

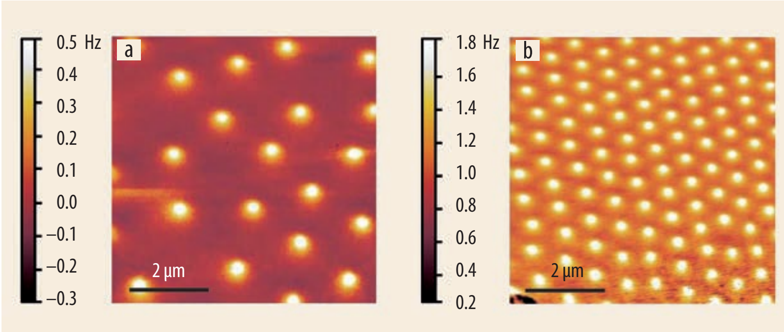

Fig. 3 MFM measurements of the vortex lattice in superconducting BSSCO at different magnetic fields: a) –1 mT, b) –4 mT. The images taken at $T = 4$ K show constant height measurements ~30 nm above the sample, with an active phase-locked loop (PLL).

is driven by a remote compressor and connected via a rotary valve that controls the frequency of the pulsing. Often, such systems are also equipped with superconducting magnets, conductively cooled by the pulse tube through thermal contacts.

This particular design ensures mechanical decoupling between the pulse tube and the sample space in such a way that the vibrations from the pulse tube cryocooler do not influence vibration-sensitive experiments, while still ensuring a good thermal contact for sufficient cooling power. Initial cooling of the whole system requires 5 – 10 hours with no magnet and 10 – 15 hours including a 9 T magnet. Usually, the cryostat and magnet are always kept cold between measurements, so that cooling down a microscope insert from $T = 300$ K to base temperature (4 K) is normally achieved within 1 – 2 hours, depending on the mass of the microscope. Warming up the microscope system also takes only about 1 hour. Thanks to the top-loading architecture, the sample space is immediately accessible after warming up. The configuration of the system allows for a turnover time of about three hours, including sample or tip exchange, which typically requires only a few minutes.

Low Vibration Sample Space

For the initial characterization, a calibrated vibration detector was used to measure vibrations in-situ directly at the sample location [1]. Careful measurements of the amplitude spectral density show a reduction of up to three orders of magnitude at the pulse tube frequency: We measure absolute vibration amplitudes below 20 nm (vertical axis) and 5 nm (horizontal axis) respectively compared to the displacements measured on the cold head itself of approximately 5 µm [2]. The measured absolute vibration amplitude along the vertical axis in the nm range is low enough to enable scanning probe microscopy measurements such as atomic and magnetic force microscopy (AFM, MFM) as described below.

Contact Mode AFM Measurements

One estimate for the overall vibrations and hence the noise encountered in a real AFM measurement is given by the relative displacement amplitude between the AFM tip and the sample without scanning.

In the simplest configuration, this quantity (z-noise) is acquired in contact mode. In contrast to most other AFMs, our instrument does not use a 4-quadrant diode detection system, but a fiber-based interferometer (**Fig. 2a**) [3]. We acquire z-height data over a few minutes (10,000 points @ 5.12 ms sample time), while keeping the tip in contact with the sample surface at a fixed lateral position. The data is line-by-line flattened to remove slow drifts below 1 Hz (e. g. piezo creep), so that the effective bandwidth is approximately 1 to 200 Hz. Typically the z-noise data has a Gaussian distribution characterized by its standard deviation (**Fig. 2b**), which in this case was $\sigma = 65$ pm rms. This value is measured with the feedback loop enabled (with the same parameters as used for regular imaging), and is almost as low as in typical liquid helium cryostats. When the feedback is turned off, the noise amplitude increases approximately by a factor of 4. To further demonstrate the low vibration noise we imaged atomically flat terraces of height corresponding to the lattice parameter $a = 0.39$ nm in our dry cryostat at $T = 3.2$ K on a strontium titanate ($SrTiO_3$) commercial wafer (**Fig. 2c, d**). This demonstrates the extremely low noise in the cryostat

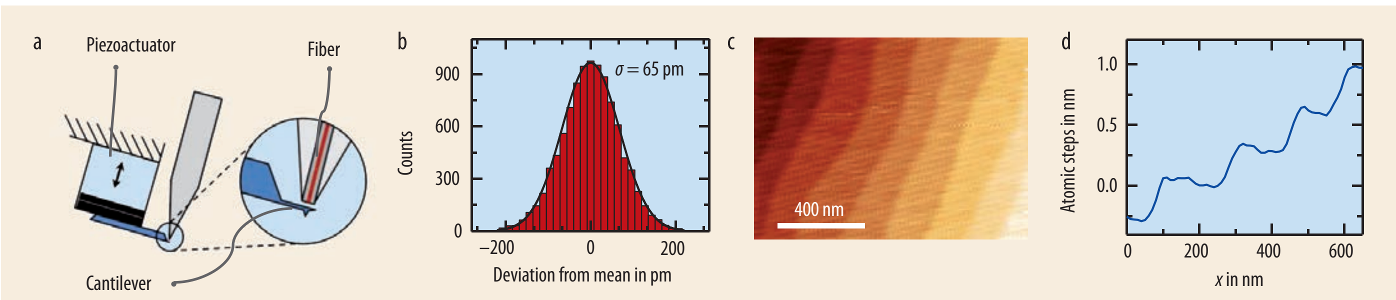

Fig. 2 a) Interferometric head for AFM measurements with built-in fiber-based interferometer. b) Contact mode noise scan histogram of the z-height values measured with a bandwidth of 1 ... 200 Hz at 3.2 K. The measured amplitude of the relative displacement standard deviation is 65 pm. c) Contact-mode AFM image of atomically flat terraces on $SrTiO_3$ (200 scan lines) at 3.2 K. The step height is 0.39 nm, corresponding to the lattice parameter of the crystal. d) Line profile showing the height of the atomically flat terraces on $SrTiO_3$.

on a real sample, hence enabling the investigation of other interesting physical phenomena further discussed below.

Magnetic Force Microscopy

Physics at low temperatures offers the challenging and rewarding opportunity of investigating fundamental properties of matter. In particular, the scanning probe family of magnetic imaging methods includes powerful techniques to probe fundamental interactions on the nanometer scale.

One interesting example of quantum objects that are intensely studied with these methods are vortices in type-II superconductors, motivated mostly by the desire to pin down the nature of high-Tc superconductivity. The magnetic properties of a superconducting vortex originate in a circular supercurrent, which allows one magnetic flux quantum ($\Phi_0 = 2.07 \cdot 10^{-15}$ Tm²) to penetrate the superconductor. The density of vortices can be tuned and is directly proportional to the external applied magnetic field in the intermediate region between the lower critical magnetic field H_{c1} and the critical field H_{c2} above which superconductivity vanishes. Through the mutual repulsion of neighboring circular currents, a vortex lattice forms in order to minimize the free energy density, which in the easiest case is hexagonal as theoretically predicted by Abrikosov [4]. However, due to natural or artificial defects present in most materials, there are additional pinning forces, which immobilize and trap vortex cores at preferred locations, distorting the ideal lattice. While vortices can move upon application of an external current and induce electrical resistance, controlled tailored pinning is desirable to reduce this adverse effect and minimize electrical losses.

Magnetic force microscopy (MFM) is a viable tool to observe such vortex lattices, e.g. on a freshly cleaved $Bi_2Sr_2CaCu_2O_{8+x}$ (Bi-2212) cuprate superconductor (**Fig. 3**). The orientation of the vortices with respect to the moment of the tip is indicated by the colour of the vortices: bright colours indicate repulsive forces. Here, the tip was scanned at the constant height of about 30 nm above the surface of a freshly cleaved piece of Bi-2212 (sample courtesy of A. Erb, TU Munich, Germany). Note that the applied field was always much lower than the coercitivity of the hard-magnetic tip (~40 mT), hence the orientation of the tip moment was kept unchanged. While at low vortex densities (**Fig. 3a**) pinning effects dominate and the lattice is disordered, higher fields and hence higher vortex densities result in a more ordered hexagonal Abrikosov pattern (**Fig. 3b**). Statistical evaluations of such measurements are often used in material science to find out about the pinning forces in these materials (see for example [5]).

As an indicator for the performance of the system, we measured the signal-to-noise ratio (SNR) from the peak heights of the isolated vortices for example at $B = 1$ mT to be larger than 20:1, with a 10 ms bandwidth, matching the SNR measured in low noise liquid helium cryostats.

Skyrmions in A Crystal

Magnetic skyrmions are nanoscale spin textures in chiral magnets. The name skyrmions allude to a non-linear field theory proposed in the context of nuclear physics by T. H. R. Skyrme [6]. In magnetic materials corresponding topologically nontrivial configurations were recently discovered within a narrow region of temperature and magnetic field [7]. The skyrmion-lattice phase can be observed in materials without inversion symmetry. The topological stability of magnetic skyrmions makes them excellent candidates for future high density magnetic storage materials [8, 9]. Skyrmions as little as 1 nm were recently reported [10]. Furthermore, the demonstration of the creation and annihilation of a single skyrmion [11, 12] shows their potential for the application in information technology.

Using a high-quality single-crystal of $Fe_{0.5}Co_{0.5}Si$ (sample courtesy of A. Bauer and C. Pfleiderer, TU Munich, Germany, [13]), both helimagnetic structures at $T = 3.2$ K in zero magnetic field as well as a skyrmion-lattice texture at

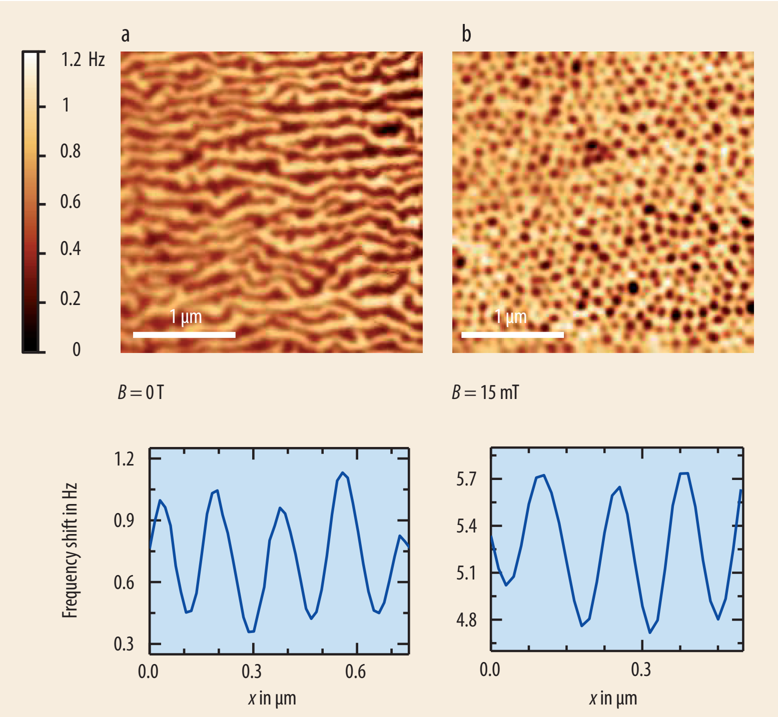

Fig. 4 MFM images of a polished surface of bulk samples of $Fe_{0.5}Co_{0.5}Si$. a) Helimagnetic phase of the sample at $T = 3.2$ K after zero-field cooling ($B = 0$ T). b) Metastable skyrmion-lattice phase measured at $T = 3.4$ K in an external magnetic field $B = 15$ mT after field-cooling.

$T = 3.4$ K in an externally applied field $B = 15$ mT (**Fig. 4**) could be observed during measurements in our labs. In both cases, the magnetic tip was kept at a constant height of 20 – 30 nm over the sample surface, with a phase-locked loop activated to track the cantilever resonance frequency. For field cooling to observe the skyrmion phase, the sample was first heated to 60 K, then the magnetic field was increased to 15 mT, and subsequently the sample was 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 and 3.6 K at the magnet.

In summary, we have shown above that our cryostat design enables very sensitive scanning probe microscopy experiments at low temperatures and in high magnetic fields. This is possible even without the need for additional spring suspension or internal damping in the microscope itself. The low noise in AFM measurements was demonstrated via imaging of atomic steps, with further applications ranging from vortex imaging in superconductors to the observation of exotic new phases in materials potentially interesting for magnetic storage at nanometer length scales.

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