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Scanned probe microscopy at millikelvin temperatures

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Abstract

Scanned probe microscopy, based on such local interactions as electron tunneling, inter-atomic forces, and near-field optics, offers uniquely powerful tools for the investigation of physical systems on the nanometer scale. Adapting these techniques to operation in the millikelvin regime will be discussed in the context of our atomic force microscope, which operates at temperatures down to 20 mK and in magnetic fields up to 9 T. The scan range at low temperatures is 4 μm $\times$ 4 μm. The instrument features a piezoelectric linear motor for vertical coarse approach, and a horizontal sample translation stage with a 2 mm $\times$ 2mm range. A fiber interferometer is used to detect the force-sensing cantilever displacement. Demonstrated performance includes the ability to detect single atomic steps on a graphite surface at 4.2 K and the ability to locate and image nanometer scale electronic devices at millikelvin temperatures.

Keywords: scanned probe microscopy, low temperature techniques, atomic force microscopy

The invention of the scanning tunneling microscope (STM) [1] has stimulated a wide array of experimental techniques, collectively known as scanned probe microscopies (SPM). Many of these techniques have been adapted for cryogenic applications, to the point that SPM experiments at liquid ^{4}He temperatures have become almost routine. There has been considerably less effort at temperatures below 1 K, however, with groups reporting operation at 0.7 K [2] and 300 mK [3].

In this paper we discuss the design and implementation of an atomic force microscope which operates down to 20 mK in fields up to 9 T [4,5]. The instrument reflects some important considerations unusual in the field of scanned probe microscopy: reliable cooling of the sample under study, extremely low power dissipation, and the ability to translate the sample over large distances in order to locate specific structures. The instrument is compact enough to fit within the bore of a high field solenoid, and is not affected by eddy current heating as the field is swept. It features a reliable coarse approach mechanism and a 4 μm $\times$ 4 μm scan range at low temperatures.

The major components of the instrument are illustrated in Fig. 1. The tip is scanned above the surface using a conventional four-quadrant piezo tube [6,7] and deflections of the force-sensing cantilever are detected with a fiber interferometer.[8] Coarse approach of the scanner unit to the surface is achieved with a piezoelectrically driven lin-

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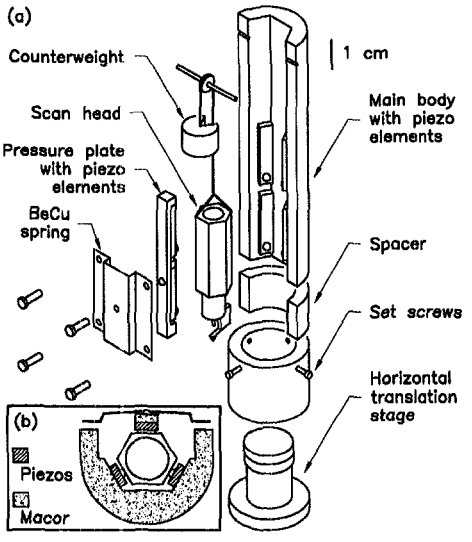

Fig. 1. (a) Exploded view of the microscope illustrating the essential components: scan head with counterweight, main body/vertical translation stage, and horizontal translation stage. (b) Cross-sectional view of the microscope showing how the scan head is held in position.

ear motor. A separate horizontal translation stage (also piezoelectrically driven) allows the sample to be moved under the tip over a 2×2 mm^2 square area. Both the linear motor and sample translation stage can be operated at low temperatures. The microscope is compact, with an overall diameter of 2.5 cm, and fits inside the mixing chamber of a dilution refrigerator/high field solenoid system. The symmetric design minimizes misalignments due to thermal contraction. The main body of the microscope was fabricated from Macor ceramic [9]. Additional parts were made from alumina and sapphire. Non-magnetic alloys were used where metal parts were unavoidable.

The microscope disassembles into three main sections: the vertical coarse approach, the sample translation stage, and the scan head. Of these elements, the vertical coarse approach is arguably the most critical, and required the greatest amount of development time. In order to avoid crashing the tip into the sample, this mechanism must be able to take steps as small as 10 nm, and yet cover distances on the order of 1 mm in a reasonable amount of time. For the instrument described here, there are the additional requirements that the microscope fit inside the bore of a solenoid, operate in high magnetic fields, and have only a very weak thermal connection to room temperature.

For these reasons, we chose to build a piezoelectrically driven linear motor operating on a "stick/slip" principle. The essential components of this system can be seen in Fig. 1. A hexagonal alumina tube containing the scanning piezotube is held by sapphire bearings on six PZT-5A piezo motor elements. Each bar-shaped element is 17.8 mm long, with a width of 2.5 mm and a thickness of 1.3 mm. One end of each bar is epoxied to the Macor body of the microscope, the other end can move freely as the bar extends or contracts under the voltage applied to the electrodes. The maximum range of motion for the bars is ± 0.3 μm at $T = 100$ mK.

Fig. 1(b) shows a cross-section of the microscope and illustrates how the hexagonal scan head is held in position. To compensate for thermal contraction, four of the bars are attached to the main body of the microscope while the remaining two are attached to an independently movable pressure plate loaded by a BeCu leaf spring. The four bearings on the microscope body are hemispherical in shape, while those on the pressure plate are flat discs (to prevent azimuthal rotation of the hexagon). An additional hemisphere on the back of the pressure plate mates with a hole in the leaf spring and ensures that the applied pressure is evenly distributed between the two bearings.

An important component of the motor design is the 8 g tungsten counterweight which balances the weight of the scan head. The weight hangs from a Kevlar thread running over a small pulley. The miniature ball bearing for the pulley was carefully cleaned of all lubricants to allow operation at low temperatures. Originally, we attempted to operate the microscope without this counterweight. Unfortunately, it would then only operate correctly over a narrow temperature dependent range of spring pressures. The correct pressure for operation at helium temperatures was too light to hold the scan head against gravity at room temperature. These

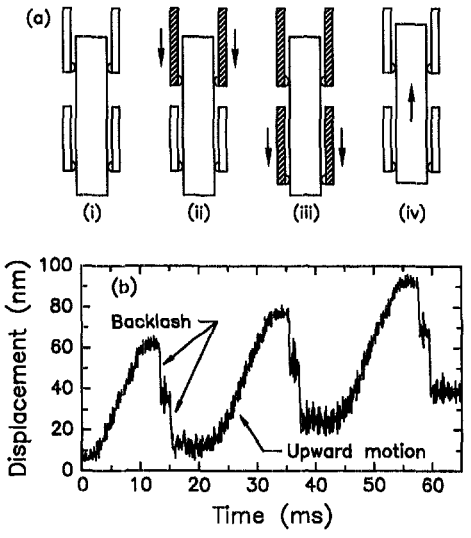

Fig. 2. (a) The principle of operation for the vertical coarse approach mechanism. (i) The stage is in the initial position. (ii) The first group of piezos is extended suddenly. (iii) The second group of piezos is extended suddenly. (iv) All the piezos are contracted slowly, bringing the stage in the upward direction. Downward motion proceeds in a similar fashion. (b) Optical fiber interferometer recording of three consecutive steps of the scanner against gravity at $T = 300$ K in vacuum.

difficulties were cured by the addition of the counterweight. Now, once the BeCu spring pressure is adjusted (by trial and error) to give reliable operation at room temperature, the motor works equally well at low temperature, and steps with and against gravity have the same size. We have not observed any instances where this arrangement has tangled or jammed upon cooling to low temperatures. Because the required spring pressure is quite light, and because the alumina hexagon has been polished to .1 μm rms roughness, we have not observed any wear on the sapphire bearings.

Operation of the motor is extremely simple. The six motor piezo elements are electrically grouped into upper and lower triads. A single step of the motor away from the surface is illustrated schematically in Fig. 2(a). First the upper and then the lower triads are extended by a step change in the applied voltage (rise time $\sim$10 μs). Then all six elements are slowly returned to their equilibrium positions. In the first phase of the step, the sudden

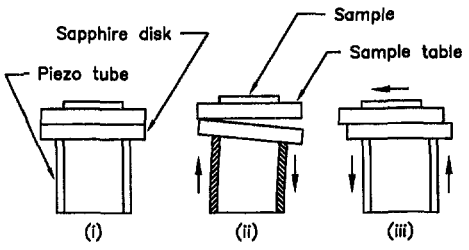

Fig. 3. The schematic principle of operation for the horizontal sample translation stage. (i) The stage in the initial position. (ii) The piezo tube is bent suddenly. (iii) The piezo tube is relaxed slowly, bringing the sample table in the desired direction.

motion causes one set of bearings to slip against the alumina hexagon while the other holds it in position. In the second phase, the motion is gentle and the bearings stick, carrying the scan head upward. Fig. 2(b) shows the typical behavior of the motor, as observed with an interferometer. The sudden extensions of each triad of piezos cause about 50 nm of backlash, but the overall progress is upward at about 15 nm per step. The smallest reliable step obtainable at $T = 100$ mK is $\approx$10 nm.

The sample translation stage, required for positioning small devices under the microscope tip, also operates on a stick/slip principle. It consists of a 14 mm diameter, 3.2 mm thick polished sapphire disk epoxied to the top of a standard piezoelectric scan tube (PZT-5A, 12.7 mm diameter, 12.7 mm length, .5 mm wall thickness). The sample is carried by a second identical disk which simply rests on top of the first, held in place by gravity. Translation of the sample is illustrated schematically in Fig. 3, and is quite similar to the operation of the vertical motor. A sharp step in voltage is applied to two quadrants of the piezo tube, with opposite polarities applied to opposite quadrants. As a result, the tube bends to one side. Because of its inertia, the disk with the sample remains approximately stationary. The voltage is then removed slowly, and the straightening tube carries the sample with it. We use a simple sawtooth waveform to drive this motion.

At millikelvin temperatures, this arrangement can move the sample in two orthogonal directions in steps as small as 10 nm or as large as 100 nm. but

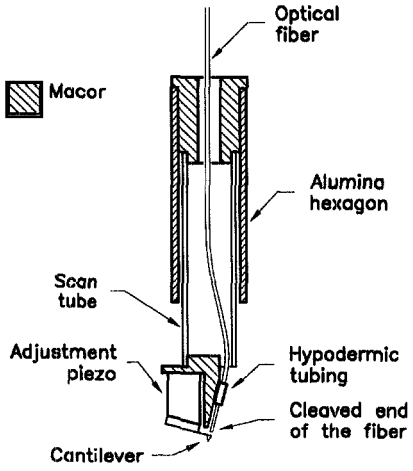

Fig. 4. Schematic illustration of the microscope scan head.

we typically use an optical microscope to approximately position the sample under the tip and limit the travel of the stage with four 000-120 screws.

Although the horizontal translation stage is quite simple, there are some mysteries associated with the design. In our original stage, the sample table rested on three sapphire hemispheres rather than a second plate. This design worked reliably for a year, and then stopped. We experimented extensively with different drive voltages and waveforms, but finally gave up and built a new, identical stage. It wouldn't work either. Eventually, the current plate-on-plate design was tried and found to translate the sample reliably, but only if the piezo tube had a large diameter. An identical stage driven by a 6 mm diameter tube would not work. If the current stage fails at some point in the future, we will likely attempt one similar in spirit to the Besoke coarse approach mechanism [10], which would also allow rotation of the sample.

The construction of the microscope scan head is illustrated in Fig. 4. We use a conventional lead-zirconate-titanate (PZT-5A) scanning tube (25 mm long, 6.4 mm diameter, .5 mm thick wall) epoxied [11] to a Macor base. Because the low temperature thermal contraction of Macor is so similar to that of PZT [12], we find no evidence of cracking or fatiguing of the scan tube, even after many thermal cycles.

We chose a fiber interferometer to detect the deflection of the force-sensing cantilever, both for simplicity of alignment, and for low power dissipation. As shown in Fig. 4, the fiber passes through a stainless steel sleeve (hypodermic tubing) mounted on the scan head. This tube provides enough friction to hold the fiber during alignment of the interferometer, while the cantilever is held in place with a small BeCu spring clip. The cleaved end of the fiber and the cantilever are manually aligned for a large fringe depth at the interferometer output. After the alignment is complete both the fiber and cantilever substrate are rigidly fastened in place with small dabs of a viscous epoxy [13]. This simple design has proven to be reliable and temperature stable.

A secondary PZT element on the end of the scan tube is used for fine control of the spacing (typically 50-100 μm) between the cantilever and the optical fiber at low temperatures. Changing the cantilever, and/or re-cleaving the fiber is done by mechanical removal of the epoxy dabs. A thin (250 μm) sapphire plate on the end of the adjustment piezo aids in this process. We use commercial Si_3N_4 cantilevers [14] without any metallic coating, although the scheme could be readily adapted to other cantilevers. Levers which are coated with metal on only one side must be avoided, since differential thermal contractions will cause severe bending and consequent misalignment of the interferometer at low temperatures.

The room temperature components of the interferometer are similar to the original design of Rugar *et al.* [8] with the addition of a neutral density filter which reduces the total power delivered at low temperatures to below 500 nW. We compensate for the reduced signal intensity by using a photomultiplier tube [15] to detect the interference signal. To avoid the complications associated with splices and fiber-to-fiber coupling, we buy standard fiber optic directional couplers (designed to operate at 633 nm) with an extra 10 m of fiber on one arm [16]. The fiber enters the vacuum space of the dilution refrigerator through a room temperature wax [17] seal, and passes into the mixing chamber via

an epoxy seal. Typically about 1 m of extra fiber is stored inside the mixing chamber close to the microscope. Once that reserve has been depleted, additional fiber is fed into the cryostat from a spool at room temperature and the two seals are re-made.

The final design and installation of this instrument was dictated by a number of unusual criteria. Foremost among these considerations was thermal contact to the sample, an issue which is of particular importance in experiments on mesoscopic systems, but which does not usually arise in conventional scanned probe microscopy. To ensure adequate cooling, we chose to mount the microscope inside the mixing chamber of our dilution refrigerator.

Care was taken to reduce and eliminate sources of noise, both mechanical and electrical. The refrigerator is suspended from a set of commercial air legs [19], and all pumping lines run through a massive concrete block before being connected to the cryostat with either flexible hose or an opposed-bellows vibration isolator [18]. The pumps themselves are mounted on a vibration isolated platform and enclosed in a soundproofed box.

As an example of the results obtainable with this instrument, Fig. 5 shows a tilt-subtracted image of the ring at a temperature of 30 mK in an applied magnetic field of 9 T. The image was taken in contact mode with an applied force of 4 nN. The 500 nW optical power used to detect the bending of the AFM cantilever did not detectably change the temperature, as measured with a RuO_2 resistance thermometer mounted close to the sample. Applying the magnetic field had no discernible effect on the performance of the microscope. There was no observable shift in the sample position relative to the microscope tip and eddy current heating effects were quite small: the sample temperature remained below 30 mK as the field was increased from zero at a rate of 0.6 T/min.

The base temperature of the system is 20 mK, and operation of the microscope in imaging mode generates negligible warming. Use of the vertical or horizontal translation stages dissipates roughly 50 μW, as deduced from the rise in base temperature of the dilution refrigerator during normal operation of these mechanisms.

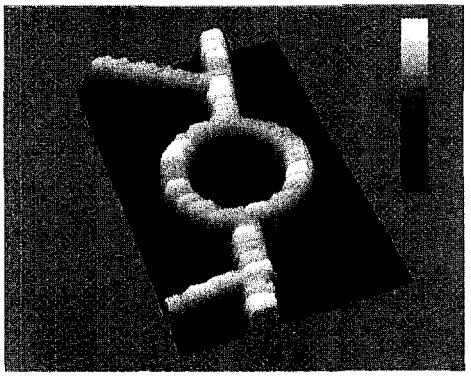

Fig. 5. Force microscope image of a mesoscopic ring taken at 30 mK in a 9 T field. The imaged area is 1.6 μm wide by 2.5 μ high. Vertical distances are indicated by the grey-scale color bar which spans 564 Å.

The noise spectrum of the microscope is white, with an rms amplitude on the order of 6 Å, resulting in a minimum force sensitivity of 6 pN, assuming the manufacturer's nominal spring constant of .01 N/m for the cantilever. Shutting off the mechanical pumps and closing the fill valve on the 1 K refrigeration stage lowers the noise by only a factor of 2, indicating that improvement will come primarily through a more rigid microscope. Our design has a rather large mechanical "loop" between the tip and the sample, and a lowest resonant frequency on the order of 700 Hz.

Although our microscope was designed for contact mode operation and uses a soft cantilever to support the tip, we investigated the oscillatory behavior of the cantilever to gain insight into how operation in a superfluid would affect "non-contact" and "tapping" microscopy modes which use vibrating tips. Cooling the microscope to 4 K in vacuum caused a $\sim$200 Hz shift in the 8 kHz resonant frequency of the cantilever, from which we estimate the stiffness of the Si_3N_4 increased by about 5%. When immersed in superfluid, however, the resonant frequency dropped to 4.32 kHz due to mass loading by the liquid. In addition, the Q of the lever dropped from $\sim$1000 down to 100, comparable to what we observe in air. Although the surrounding

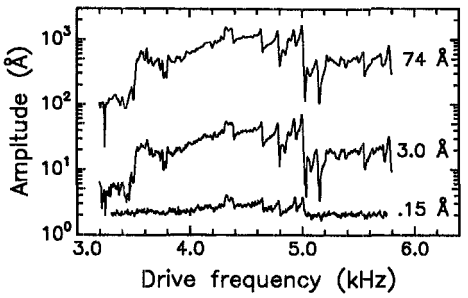

Fig. 6. Amplitude response (peak-to-peak) of the driven cantilever at 40 mK inside the dilution refrigerator mixing chamber. The approximate peak-to-peak amplitude at the base of the cantilever (obtained by driving the z motion of the scan tube) is given for each curve. The peak at 4.32 kHz, visible in all three curves, is the cantilever resonance.

^{3}He-^{4}He solution is a superfluid, it has a viscosity $\eta T^2 \sim .3$ μpoise K^2 below 100 mK [20].

More importantly, we find that the power spectrum for the tip motion is very complex, even for very low drive amplitudes. This behavior, illustrated in Fig. 6, is similar to what is observed in tapping mode microscopy in normal liquids, and is attributed to the excitation of acoustic modes in the fluid [21]. Although we have not yet attempted it, we expect that tapping mode microscopy should also be possible with our instrument. The phase response of the cantilever, however, is extremely flat in the region of the resonance, which will likely rule out other forms of non-contact microscopy. On the other hand, all of our investigations have been on V-shaped cantilevers with very low spring constants. It is possible that the use of stiffer, straight rectangular levers, or levers with a round cross-section [22] may improve the situation. This is an area worthy of additional study.

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