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Scanning Probe Manipulation of Magnetism at the $LaAlO_3/SrTiO_3$ Heterointerface

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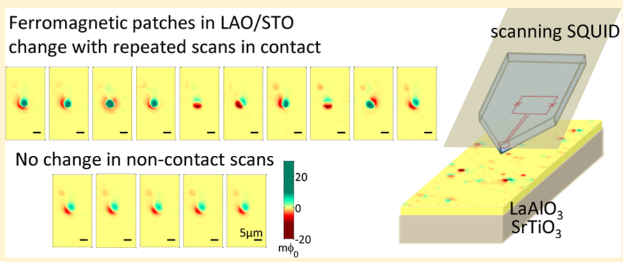
ABSTRACT: Manipulation of magnetism is a longstanding goal of research in exotic materials. In this work, we demonstrate that the small ferromagnetic patches in $LaAlO_3/SrTiO_3$ heterostructures can be dramatically changed by in situ contact of a scanning probe. Our results provide a platform for manipulation of small magnets through either a strong magneto-elastic coupling or sensitivity to surface modification. The ability to locally control magnetism is particularly interesting due to the presence of superconductivity with strong spin–orbit coupling in $LaAlO_3/SrTiO_3$.



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$LaAlO_3$ (LAO) and $SrTiO_3$ (STO) are both nonmagnetic insulating materials, but the interface between the materials exhibits conductivity[1,2] and magnetism[3−8] above a critical thickness of the LAO.[9,10] It has been shown that stress affects conductivity in global transport measurements in LAO/STO,[11] and it is generally known that stress can change the magnetization of a material (the inverse magnetostrictive effect). Physical contact with a sample can affect magnetism by stress-related change of the crystalline anisotropy[11] and also by surface modifications such as transfer of charge or adsorbates, which are both known to affect conductivity in LAO/STO.[12−14]

In this work, we show that the ferromagnetic islands in LAO/STO are very sensitive to in situ contact with the sample. We contact the surface of the sample with the tip of a scanning superconducting quantum interference device (SQUID) probe, and simultaneously image the magnetic landscape. Both the moment and orientation of the ferromagnetic patches changed when the tip was in contact and then remained stable until further contact. Contact experiments with a large number of repetitions revealed an underlying anisotropy along crystalline axes, STO miscut terraces, or other underlying order. These observations imply that each ferromagnetic region consists of submicrometer ferromagnetic domains that change configuration due to stress and provide experimental input to guide hypotheses of the nature of magnetism in LAO/STO. It is also possible that contact modifies the surface charge and/or adsorbates.

LAO/STO samples were prepared by growing 3−15 unit cells (uc) of LAO on TiO_2-terminated {001} STO substrates, as previously described.[10] Structural, interface, and surface characterization of similar samples have also been previously described.[15,16] Our measurements were performed with two scanning SQUID microscopes with base temperatures of 4 K and 18 mK.[17,18] Each SQUID has a 3 μm pickup loop that records the change in magnetic flux as a function of position as the device is scanned over the sample surface in units of the flux quantum, Φ_0. Our SQUID probes detect the local changes in magnetic flux, rather than the magnitude of ambient flux. When analyzing our data we define our zero as the measured signal on a nonmagnetic sample or away from any sample. The SQUID is fabricated on a silicon chip that is polished to a corner and mounted on a cantilever to bring the pickup loop close to the sample surface (Figure 1a). When the pickup loop is scanned over a magnetic dipole (or a small bar magnet), it captures the field generated by the dipole at different locations. From such field maps we determine the presence of the dipole, its moment, and orientation,[7,10] The SQUID can detect nanoscale ferromagnetic objects with moments as small as $10^4\ \mu_B$ (in dc; $\sim 10^2\ \mu_B/\sqrt{\mathrm{Hz}}$ in ac) even though they are much smaller than the physical size of the pickup loop. By pushing the cantilever into the sample, the SQUID tip applies a force of $\sim 1\ \mu$N (Figure 1b), producing a local stress that decays as the square root of the distance from the contact point. We expect these forces to be well within the mechanically elastic regime.

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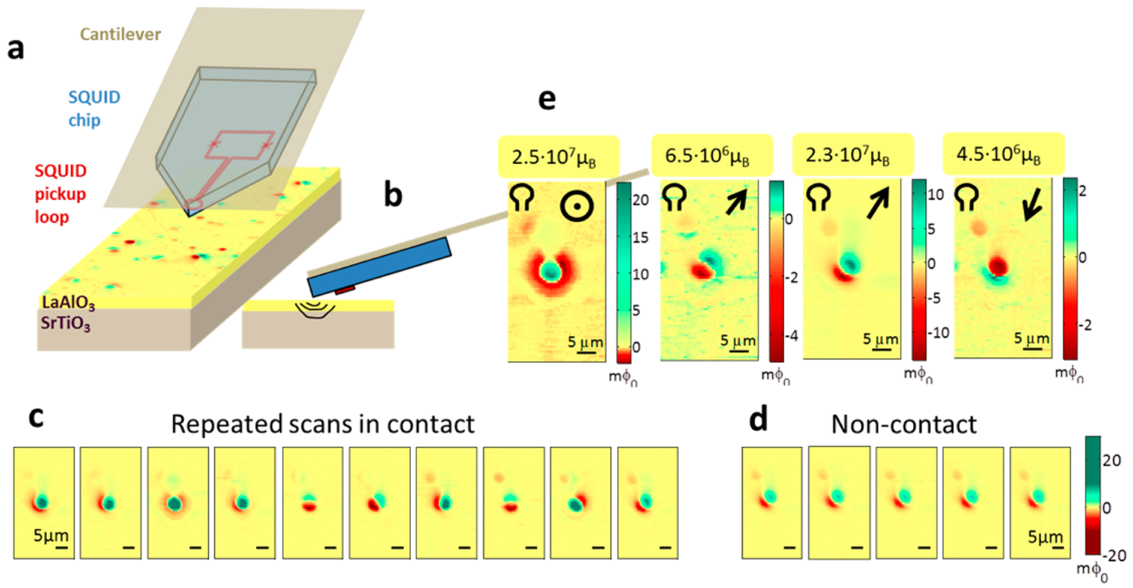

Figure 1. Ferromagnetic patches change with local contact applied by the SQUID's tip. (a) Cartoon of the measurement configuration. (b) The SQUID chip is mounted on a cantilever and can be pushed onto the sample surface during a scan. The sensing area is ∼7 μm away and 1 μm above the contact point. (c) Ten successive scans (4 K; 15 uc sample) of a 25 μm × 45 μm area reveal changes in a ferromagnetic patch. The tip is out of contact between scans and brought into contact at the start of the next scan. (d) Five successive scans obtained directly after the scans in (c) but without contact with the sample surface (1 μm higher than in (c)) show no change in the patch. (e) Four scans in contact plotted on the full flux scale show the change in orientation and moment (noted above each scan). The small arrow on each scan marks the orientation angle of the dipole, which is extracted directly from the data using the magnitude and orientation of the dipole. A weaker ferromagnetic patch in the scanned region follows the same behavior, although the orientation of the two patches in the scans do not appear to be related.

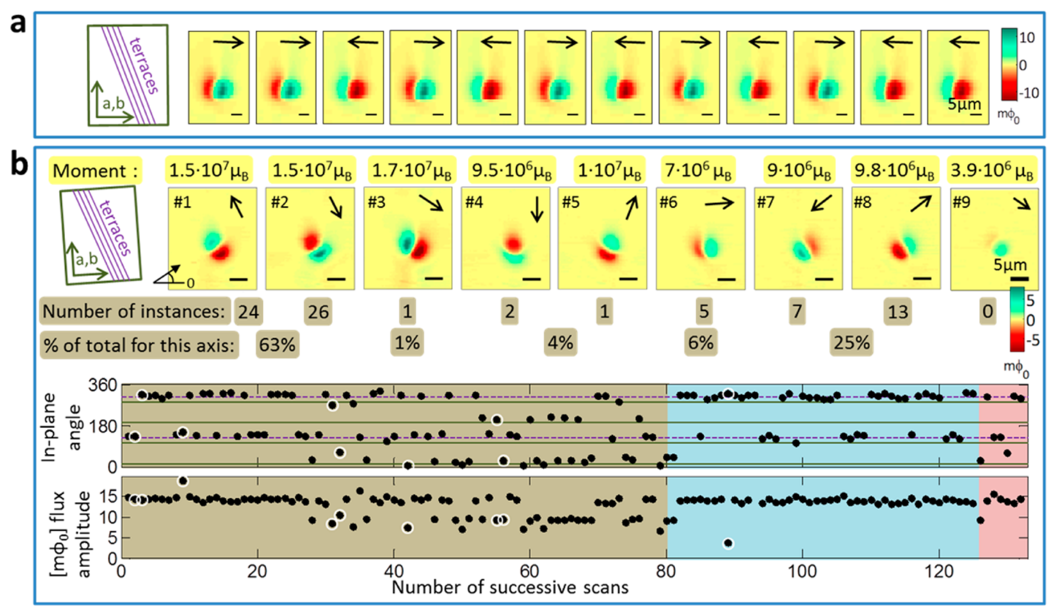

Figure 2. Magnetism scanned in contact aligns in preferred orientations. (a) Twelve successive scans in contact of a ferromagnetic patch in the 15 uc sample at 4 K. The sketch indicates the crystallographic directions a,b of the STO and the orientation of the terraces (determined by atomic force microscopy). (b) Nine orientations (yellow background) observed for a different ferromagnetic patch on the same 15 uc sample in a different cool-down. The scans are a subset of a sequence of 132 scans in contact. All other scans in this sequence repeat one of these arrangements. For each scan, we specify the moment, the number of identical scans, and the percent of the total for each axis, observed in the first 79 scans (brown background). The lower panel shows the in-plane orientation angle and the flux amplitude (proportional to the moment) as a function of the number of successive scans in contact. A white circle around a data point indicates that the scan was plotted in the top panels of (b); the circles appear in the same order as the images. The solid green lines in the in-plane angle panel mark the crystallographic direction, and the dashed purple lines mark the terrace orientation. The first 79 scans (brown background) were taken with a tip−sample force of ∼2 μN. Scans 80−125 (blue background) were taken in contact with a force of ∼0.5 μN. At this lower force, we found that most of the orientations (43/46) were #1 and #2. The last part of the sequence (red background) was taken with a force of ∼3 μN. More orientations become available at 3 μN.

The ferromagnetic patches dramatically change when scanned in contact (Figure 1c). When there was no contact during the scans, no change in the patches was detected (Figure 1d, SQUID tip is 1 μm above the surface). We repeated this sequence of 15 scans (Figure 1c,d) five times and found consistent behavior. Figure 1e illustrates four instances of scans in contact, recording changes in both the magnitude of the moment and its orientation. The characteristic size of the dipole features is due to a convolution of the field generated by each ferromagnetic patch with the shape of the SQUID's pickup loop (circle with leads, sketched on each scan). Using the recorded flux data and the known field distribution near a magnetic dipole, we determined that the moment of the ferromagnetic in Figure 1e changes over approximately 1 order of magnitude from 2.6×10^6 to 3.6×10^7 μ_B. The as-cooled orientation had the highest moment. The moment is linear with the amplitude of the measured flux (at a given height), and thus we describe the moment in terms of flux amplitude (mΦ_0) to maintain the closest relation to the raw data in our analysis.

The sensitivity to tip−sample contact varied between patches. We repeatedly scanned (in and out of contact) 238 isolated ferromagnetic patches from 12 samples of various LAO thickness. We found that none of the patches changed when scanned out of contact but scanning in contact changed 55% of the patches. To rule out experimental variations (e.g., of the applied stress) as the cause of this variability, we scanned in-contact over a relatively large region (60 μm × 80 μm) on a 10 uc LAO sample in which the density of ferromagnetic patches was relatively high.[10] In this case, all of the ferromagnetic patches in this region were affected by the same scan. For 20 repeated scans in contact, 70% of the dipoles changed (10 total) between successive scans. The change in the moment was different for each ferromagnetic patch, ranging from a patch with mean moment of 6×10^7 μ_B with standard deviation of 7×10^6 μ_B (12%) to a patch with mean moment of 1×10^7 μ_B with standard deviation of 4×10^6 μ_B (40%). There was no apparent relationship between the sensitivity to contact and the magnitude of the moment or the location in the scanned area.

Throughout our measurements, most of the ferromagnetic patches that responded to contact with the SQUID were tested with a small number of repeated contact scans (3−20), and the changes in their orientations seemed random. However, we rigorously examined four ferromagnetic patches by taking a larger number of contact scans, which allowed us to build up clearer statistics. From these sets we found preferred moment orientations and a relation between the moment and the orientation.

Figure 2 describes our observations on two individual ferromagnetic patches that were scanned in contact approximately 100 times. One patch alternated between the two orientations shown in Figure 2a, both with moment of $\sim 7 \times 10^7$ μ_B. The two orientations defined an anisotropy axis; the patch exhibited nearly equal probabilities of alignment or antialignment to this axis (45 and 55% of 95 instances, respectively). This axis was parallel to one of the in-plane crystallographic directions (error in crystallographic direction, ∼4°). The other patch presented more complicated behavior. Only two orientations (#1 and #2 in Figure 2b) were detected in 27 out of the first 28 scans of this patch, defining an anisotropy axis in the direction of the miscut terraces of the substrate. Similar to the patch in Figure 2a, this patch exhibited equal probabilities of alignment or antialignment with this axis. After 28 scans new orientations were detected, but the original anisotropy axis continued to reflect the preferred orientations (75% of the 132 scans in this sequence). Another less probable axis (defined by orientations #7 and #8 in Figure 2b) was found in 25% of the instances in this sequence and in 17% of the total experiments. The remaining orientations (#3−5, #8, #9) appeared with much lower frequencies (1, 4, and 6%).

Though suggestive, it is still not clear that all ferromagnetic patches have an anisotropy axis. For example, the patch described in Figure 1c−e did not show preferred orientations even after 50 scans in contact. Yet some of the orientations repeated, suggesting that the number of scans was insufficient compared to the number of preferred orientations.

The moment and orientation of each patch were correlated with each orientation fixed to a given moment. Different orientations could have the same moment. We noted a typical pattern for the evolution of the moment. Most orientations that appeared after repeated contact scans had a lower moment than the as-cooled orientations (Figure 2b). We observed this trend in patches in the 15, 10, and 5 uc samples. In one extreme case, repeated scans with a force ∼3 μN on the 5 uc sample reduced the moment of a single patch by 2 orders of magnitude from 7×10^7 μ_B to below our detection threshold (a few 10^5 μ_B in that cooldown). This behavior supports a scenario in which each ferromagnetic patch is composed of small domains that change their configuration when the tip contacts the sample.

We also changed the force applied by the cantilever (Figure 2b). Stronger contact encouraged the appearance of new and less likely orientations, while weaker contact caused less-dramatic changes in the orientation and favored the orientations along the anisotropy axes. The force dependence data is limited due to the small force range that we can apply by pushing the cantilever into the sample.

Finally, we demonstrated the use of tip−sample contact for control of the magnetic landscape. We made contact with the SQUID to modify the magnetism in a specific area of the sample (Figure 3). The change in magnetism did not relax after the contact between the SQUID and the sample was broken. Modified patches remained stable up to 3 months at 0.5 K and up to one week at 4 K, as long as further scans were conducted without contact.

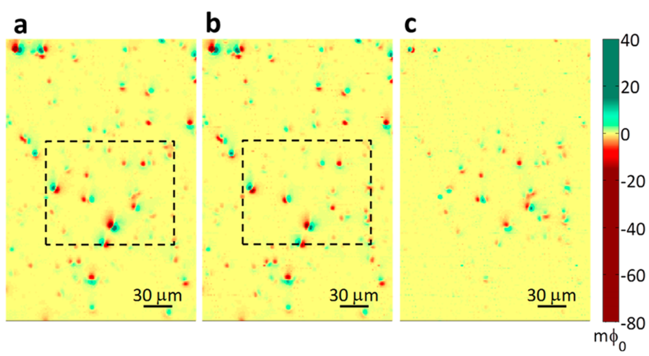

Figure 3. Control of magnetism. (a) A scan without contact between the tip and the sample (10 uc, 4 K), showing the as-cooled distribution of ferromagnetic patches. Following this scan, a subregion (dashed square) that includes ∼20 ferromagnetic patches was scanned in contact. (b) A later noncontact scan of the full range shown in (a). Only patches in the subregion that was scanned in-contact are different than in (a). (c) The difference between (a) and (b). A square that was "written" in the subregion that was scanned in contact is visible and remained stable afterward.

In addition to physical contact we considered a few other possible interactions between the SQUID and the sample. The main candidate effects are the current in the SQUID's pickup loop and field coil, and the electrostatic excitation of the cantilever. We found that the magnitudes of the current through the pickup loop and field coil do not influence the ferromagnetic patches, and that magnetic manipulation occurred even when no current was applied to both. We also followed the magnetometry signal as we gradually bring the SQUID closer to the sample and found that the effect of manipulation appears suddenly as we contact the surface and does not have a gradual onset. Any fields produced by the SQUID are not only quite small, but also change slowly compared to the onset of contact. The magnetic fields in the center of the SQUID loops for the largest currents we applied are 0.25 and 2 G for the pickup loop and field coil respectively. Half a micrometer away from the center is falls by 8 and 0.5%, respectively. As we bring the SQUID closer to the sample our resolution for the steplike appearance of the manipulation effect is about 0.2 μm. Consequently, we conclude that physical contact is causing the manipulation effect.

Strain resulting from an applied stress can change spacing between atoms and bond angles and result in a change in anisotropy.[19] Our observations, mainly the variability in the response to strain and the different outcomes for scans with nominally identical stress, imply that there is a source of disorder that competes with the effect of contact. Sources of disorder may be the relationship between the orientation and shape of the patch and the STO terraces, distribution of surface charges, or oxygen vacancies. This hypothesis is consistent with our previous observations of sample-to-sample variability and an inhomogeneous distribution of the ferromagnetic patches.[7,10] Another candidate scenario involves the manipulation of surface charges and/or adsorbates, which are known to play important role in the conductivity of LAO/STO.[12−14] Although the relation between surface effects and magnetism remains unknown, magnetism is related to the polar interface by its critical thickness.[10]

The change in moment observed in repeated scans in contact of a certain ferromagnetic patch, and the correlation between the orientation and the magnitude of the moment suggests that each ferromagnetic patch consists of several smaller ferromagnetic domains. Reorientation of these domains with stress or other contact-related effect changes the net external flux. Since these individual domains are below our spatial resolution, we measure only the total moment of each domain configuration. The preferred anisotropy axes may be due to the underlying crystalline anisotropy, the shape of a patch, or another source of directionality like the STO terraces that could serve as pinning sites for domain walls.

Theoretical work by Michaeli et al.[20] as well as by Kampf et al.[21] suggest the possibility of an exotic superconducting state at this interface, which is believed to have meaningful spin−orbit coupling.[22,23] We speculate that the ability to locally control magnetism may be particularly interesting in the context of this superconductivity. More work is needed to understand the relationship between magnetism and superconductivity in this system.

In addition, more work is needed to understand the origin of the magnetism. Any correct theoretical model of this surprising magnetism must be able to explain the phenomena observed here.

In summary, we found that isolated ferromagnetic patches at LAO/STO interfaces are sensitive to local contact between the SQUID's tip and the sample, which causes stress or modifies the surface. The patches respond to contact by switching between states with different orientations and moment magnitudes. The final state is stable after removal of contact. This stability demonstrates our ability to control magnetism in this system. We detected an underlying anisotropy along crystalline axes, STO miscut terraces, or other sources of directionality. In addition to stress-related domain-breaking, we consider the possibility of contact affecting magnetism by manipulating surface charges and absorbates.

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Notes

The authors declare no competing financial interest.

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