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Coexistence of Magnetic Order and Two-dimensional Superconductivity at $LaAlO_3/SrTiO_3$ Interfaces

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A two dimensional electronic system forms at the interface between the band insulators[1,2] $LaAlO_3$ and $SrTiO_3$. Samples fabricated until now have been found to be either magnetic or superconducting, depending on growth conditions[3,4]. Combining high-resolution magnetic torque magnetometry and transport measurements, we report here magnetization measurements providing direct evidence of magnetic ordering of the two-dimensional electron liquid at the interface. The magnetic ordering exists from well below the superconducting transition to up to 200 K, and is characterized by an in-plane magnetic moment. Surprisingly, despite the presence of this magnetic ordering, the interface superconducts below 120 mK. This is unusual because conventional superconductivity rarely exists in magnetically ordered metals[5,6]. Our results suggest that there is either phase separation or coexistence between magnetic and superconducting states. The coexistence scenario would point to an unconventional superconducting phase as ground state.

Superconductivity and magnetic order are in general mutually exclusive phenomena. Nonetheless, the coexistence of magnetism and superconductivity has been suggested for finite momentum pairing states[5,6]. Coexistence of magnetism and superconductivity has been reported in a few 3D superconducting systems[7-9], such as $RuSr_2GdCu_2O_8$ and UGe_2. The question remains if such coexistence can occur in a two-dimensional electronic system. An intriguing candidate is the interface between the two band insulators $LaAlO_3$ (LAO) and $SrTiO_3$ (STO). At their *n*-type interface a conducting two-dimensional electron liquid is generated. Moreover, the LAO/STO interface was also reported to have a 2D superconducting ground state[3].

For this system, magnetic ordering was suggested[4] by Brinkman *et al*., who deduced the presence of magnetic scattering centers from the temperature dependence of the interface resistance R and a hysteresis of R during the sweep of magnetic field H. Different magnetotransport studies indicate an antiferromagnetic order[10] or a non-uniform field-induced magnetization and strong magnetic anisotropy[11]. Recently, it was found that at both chemically treated STO bulk and LAO/STO interfaces, charges are electronically phase separated into regions containing either a quasi-two-dimensional electron gas phase, a ferromagnetic phase persisting above room temperature, or a diamagnetic/paramagnetic phase[12] below 60 K. On the theoretical side, electronic structure calculations yield complicated pictures for the agnetism at the interface layers[13–16]. Specifically, the calculations do not support magnetically ordered moments at the interface of LAO/STO bilayer covered by vacuum[17]. Consequently, any observed magnetism must originate from strong electronic correlations.

Coexistence of magnetism and superconductivity has not been reported at the LAO/STO interfaces. The ground state was found to be controlled by growth conditions, carrier concentration[18], and external magnetic field[19]. These experimental observations based on transport properties suggest that the two phenomena do not coexist (see, *e.g.*, Fig. 16 of Ref.18).

To clarify this issue, we have grown LAO/STO interfaces, measured their superconducting properties by transport measurements, and then applied cantilever-based torque magnetometry as an extremely sensitive and direct method to measure a possible magnetic moment m of the sample.

Torque magnetometry directly determines m by measuring the torque τ of the sample mounted on a cantilever in an external magnetic field H. As the torque is given by $\tau = \mathbf{m}\times\mathbf{B}$, the method detects the component of **m** oriented perpendicular to **B**. Due to its great sensitivity, this method has been applied to determine the magnetic susceptibility of very small samples, to analyze tiny magnetic signals, and, in some cases, even to accurately map Fermi surfaces[20–22].

In our setup, τ was measured with the sample glued to the tip of a 25 μm or 50 μm thick cantilever. H was applied at a tilt angle ϕ with respect to the c-axis (perpendicular to the interface). The cantilever deflection was detected capacitively. The moment m is given by $m = \tau/(\mu_0 H \sin\theta)$, where μ_0 is the vacuum permeability, and θ is the angle between m and H (with m in plane, $\theta = 90^{o} - \phi$, see discussion below). We used the measured angular dependence of the zero-field capacitance of the cantilever setup to calibrate the spring constant of the cantilever. Knowing the spring constant, we quantitatively determine the value of m. The cantilever setup can resolve changes[22] in m of $\delta m = 10^{-13}$ - 10^{-12} Am^2 at 10 T.

All samples investigated were grown using nominally identical parameters for the substrate preparation and the pulsed laser deposition. The films were patterned with Nb ohmic contacts and painted with silver paste on the back. The only intended difference between the samples is that for one reference sample (named "0 u.c." sample), a shutter in front of the substrate was used to block the growth of LAO (Fig. 1(a)). The resistance of the interface samples was measured using the Nb ohmic contacts. The LAO/STO interfaces were found to be superconducting below 120 mK. The superconducting temperature is slightly lower than that of many other LAO/STO samples grown in the same condition, which might be the result of unintended variations of growth parameters.

An example of the $\tau - H$ dependence is shown as the red curve in Fig. 1(b) for a 5 u.c. sample. The torque signal has a pronounced reversible curve with a sharp "cusp" at low field. This cusp is displayed clearly by Fig. 1(c), which zooms into this cusp. Fig. 1(d) shows m determined from the $\tau - H$ curve at -2 T $\leq \mu_0 H \leq$ 2 T. The V-shape of the $\tau - H$ curve centered at H =0 yields a nonzero, H-independent m for $\mu_0 H$ up to 0.5 T. Close to $H = 0$, m jumps to 5×10^{-10} Am^2, corresponding to 0.3 ~ 0.4 μ_B per interface unit cell (assuming that the signal is generated by the STO unit cell next to the interface, see below). The values of m very close to zero field ($|\mu_0 H| \leq 5$ mT) are hard to determine, because the small H causes a large relative noise in m. At $|\mu_0 H|$ = 5 mT, $\delta m \sim 4 \times 10^{-10}$ Am^2, which is close to the magnitude of m. Starting at fields of order 1 Tesla, m diminishes gradually at higher H, suggesting that an additional contribution appears in high fields. This high-field contribution was found to vary among different runs. Below we focus on the low-field behavior.

To explore whether the torque signals originate from the LAO/STO interface, we performed control experiments using reference samples. Sizable torque signals were only observed from samples containing LAO/STO interfaces, the torque of which exceeds that of all background samples by two orders of magnitude (Fig 1(b)). In particular, the superconducting Nb ohmic contacts are unlikely the source of the torque signal, as the torque is found far above the upper critical field of Nb (0.4 T for bulk or 2 T for thin films at 0 K). Moreover, all background m will be oriented closely parallel to H, thus creating small torque responses only. The background m is also proportional to H, as these materials are paramagnetic or diamagnetic. Furthermore, we measured a 5 u.c. thick LAO film grown on a LAO substrate. The torque signal is again two orders of magnitude smaller than that of the 5 u.c. LAO/STO sample, excluding the possible contribution from defects in the LAO film (see supplement). We therefore conclude that the observed large torque indeed arises from the presence of the LAO/STO interface.

A chief motivation for our study was to determine whether the superconductivity and magnetic order appear simultaneously or exist as separate phases in the $T - H$ phase diagram. We observe that below the superconducting T_c, the magnetic ordering signal and the superconducting state coexist. For the sample of Fig. 2, for example, the superconducting transition occurs at 120 mK at H = 0, with a resistance foot extending to 25 mK. The $R - H$ curves measured at 20 mK with H parallel and perpendicular to the interface plane are plotted in Fig. 2(b). While the interface is superconducting, the $m - H$ curve at 20 mK displays the same jump at small fields (Fig. 2(c)) as that observed at higher temperatures (Fig. 1(d)). Notably, a finite m is recorded at $\mu_0 H \sim$ 5 mT, while the sample resistance R does not reach the normal-state value until $\mu_0 H \sim$ 20 mT. The magnetic ordering signal and the superconducting state are therefore found to coexist.

The magnetic ordering signal is robust at elevated temperatures. For the 5 u.c. LAO/STO sample, m does not show significant temperature dependence even up to 40 K (Fig. 3), the highest T at which this sample was investigated. In another 5 u.c. LAO/STO sample, m was found to be nonzero up to 200 K (see supplement). Such T - dependence is consistent with previous results[12], reporting the existence of an ordering state at room temperature. The high magnetic ordering temperature indicates a strong magnetic exchange coupling.

The magnetic field dependence of m can be described by the Langevin-function characteristic for superparamagnetism, where spins are aligned in small-size domains to behave as large classical magnetic moments[23]. However, superparamagnetic samples usually show a strong temperature dependence in the low-field $m - H$ curves, a feature missing in the $m - H$ curves in Fig. 3. Noise in our measurements of m at fields close to zero may obscure this feature. Because m saturates at about 30 mT at T up to at least 40 K, the lower bound of the collective classical moment is around $10^3\ \mu_B$. On the other hand, the $m - H$ curves are also consistent with a very soft ferromagnet whose hysteresis loop is hidden by the m noise at $|\mu_0 H| < 5$ mT. Although these two possibilities cannot be distinguished by our data, all of them suggest a strong ferromagnetic-like magnetic coupling within domains.

To determine the orientation of the magnetic moment, we performed a series of torque measurements in which the sample tilt angle was varied (see inset of Fig. 1(b)). Because $\tau = \mathbf{m} \times \mathbf{B} = \mu_0 m H_\perp$, where $H_\perp$ is the component of H perpendicular to $\mathbf{m}$, the orientation of the moments can be discerned by tracking the angular dependence of the torque signal. In highly anisotropic system, m is determined by $H_\parallel$, the field component parallel to m. Thus if $H_\parallel$ is large enough to saturate m, τ will increase as a sine function of the angle between H and m. On the other hand, once $H_\parallel$ is insufficient to saturate m, τ will stop following the sine behavior.

The angle-dependence shows that the saturation magnetic moment stays in the plane of the interface. We carried out low-field torque measurements at 300 mK at 30 different tilt angles. Fig. 4 shows the $\tau - H$ curves at several selected angles ϕ. As shown in Fig. 4(a), as ϕ changes from 15^{o} to 94^{o}, τ decreases monotonically and slowly approaches zero at $\phi = 90^{\circ}$, where **H** is almost parallel to **m**. On the other hand, as ϕ varies between $+15^{\circ}$ and -10°, **H** is almost perpendicular to **m**. $H_{\parallel}$ decreases and eventually changes to the opposite direction. The in-plane magnetic moment drops to zero once $H_{\parallel}$ is close to zero. As a result, the $\tau(H)$ curves swing from a positive saturation at $\phi \sim 15^{\circ}$ to a negative saturation at $\phi \sim -10^{\circ}$.

Our data show that 2D-superconductivity and magnetic order coexist at *n*-type LAO/STO interfaces. The results leave the question open whether the same electrons are generating the superconducting and the magnetic order. The measured results can be accounted for by scenarios of spatial phase separation, in which inhomogeneous magnetic and superconducting electron layers are generated either in different lateral puddles, or at different depths away from the interface. One possible cause of such inhomogeneities is a non-uniform distribution of possible oxygen vacancies in the STO. This notion is in accord with a proposal that the oxygen vacancies in the interfacial TiO_2 layers stabilize ferromagnetic-type order of the Ti ions close to the interface, as supported by DFT-calculations[24]. In this scenario the superconducting phase is in close contact to the ferromagnetic phase, so the superconducting phase is affected by the ferromagnetism. Furthermore, the data are also consistent with the idea that the same electron system forms a magnetically ordered, superconducting electron liquid.

We note that, after our submission of the manuscript[25], two experiments were reported to support the coexistence of superconductivity and ferromagnetism at LAO/STO interface, based on hysteretic magnetoresistance[26, 27] and scanning SQUID imaging[28].

In conclusion, using torque magnetometry we have performed quantitative measurements of the magnetic moment of LAO-STO interfaces at wide-range magnetic field and broad temperature range, directly showing the presence of magnetic order in the two-dimensional electron liquid of LAO/STO interfaces. The order is characterized by a superparamagnetic-like behavior, with saturation magnetic moments of ~ 0.3 μ_B per interface unit cell oriented in-plane, persisting beyond 200 K. Below 120 mK, the ferromagnetic-like magnetic order and the 2D-superconductivity are coexisting.

Materials and Methods

The $LaAlO_3/SrTiO_3$ heterostructures were grown at the University of Augsburg using pulsed-laser deposition with *in-situ* monitoring of the $LaAlO_3$ layer thickness by reflection high-energy electron diffraction. The single crystalline $SrTiO_3$ substrates were TiO_2 terminated. Their lateral size is 5×5 mm^2 and their thickness is 1 mm. The $LaAlO_3$ layers were grown at an oxygen pressure of 8×10^{-5} mbar at 780 ^{o}C to a thickness of 5 u.c with a subsequent cooldown to 300 K in 0.5 bar of oxygen. The sputtered ohmic Nb contacts filled holes patterned by etching with an Ar ion-beam. The reference (0 u.c.) samples were grown in the same conditions (oxygen pressure of 8×10^{-5} mbar at 780 ^{o}C).

The magnetization measurements were preformed with a home-built cantilever-based torque magnetometry apparatus at MIT. Cantilevers are made from thin gold or brass foils. We deposit gold film on a sapphire and put it under the cantilever. The torque is tracked by measuring the capacitance between the cantilever and the gold film, using a GR1615 capacitance bridge or an AH2700A capacitance bridge. To calibrate the spring constant of the cantilever, we rotate the cantilever setup under zero magnetic field to measure the capacitance change caused by the weight of the sample wafer.

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Author contributions

The studies were designed, planned and analyzed by all authors, who also wrote the manuscript. C. R. grew the samples, L. L. carried out the torque and resistivity measurements and the data analysis. Correspondence and requests for materials should be addressed to R.A. (ashoori@mit.edu).

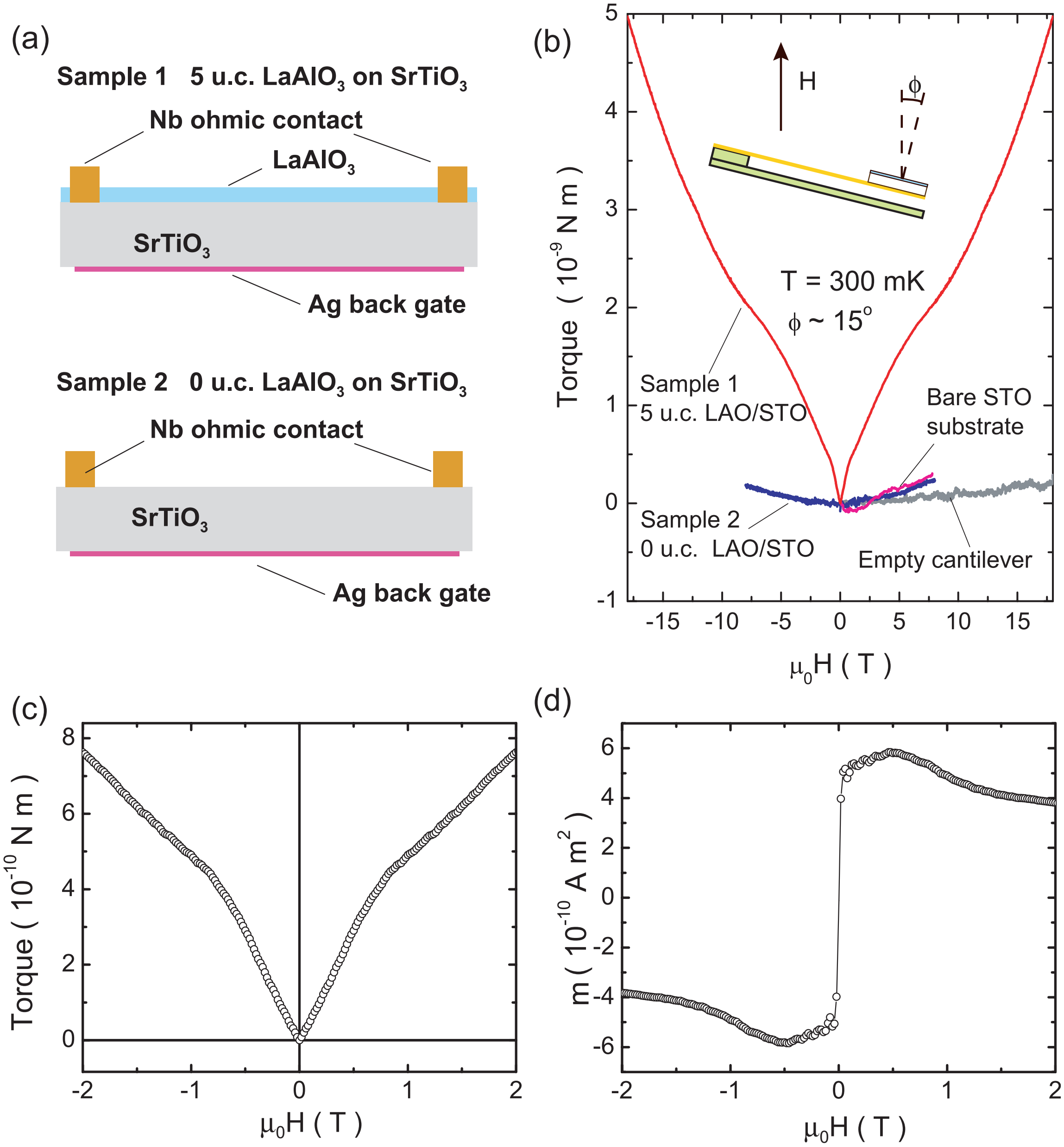

Fig. 1 **Torque magnetometry of oxide interface LAO/STO.** (Panel a) The schematics of an interface sample (Sample 1) and of a 0 u.c. background sample (Sample 2), which were grown in the same conditions. (Panel b) The field dependence of the torque curves of various test samples (cantilever only, bare STO substrate, and the 0 u.c. sample) and a interface sample, taken at $T = 300$ mK and tilt angle $\phi \sim 15^{\circ}$. The inset shows a schematic of the cantilever setup. (Panel c) In Sample 1, a field dependence of the torque curve is linear and symmetric below 0.5 T. (Panel d) In Sample 1, the magnetic moment m jumps to a finite value within mT near zero field.

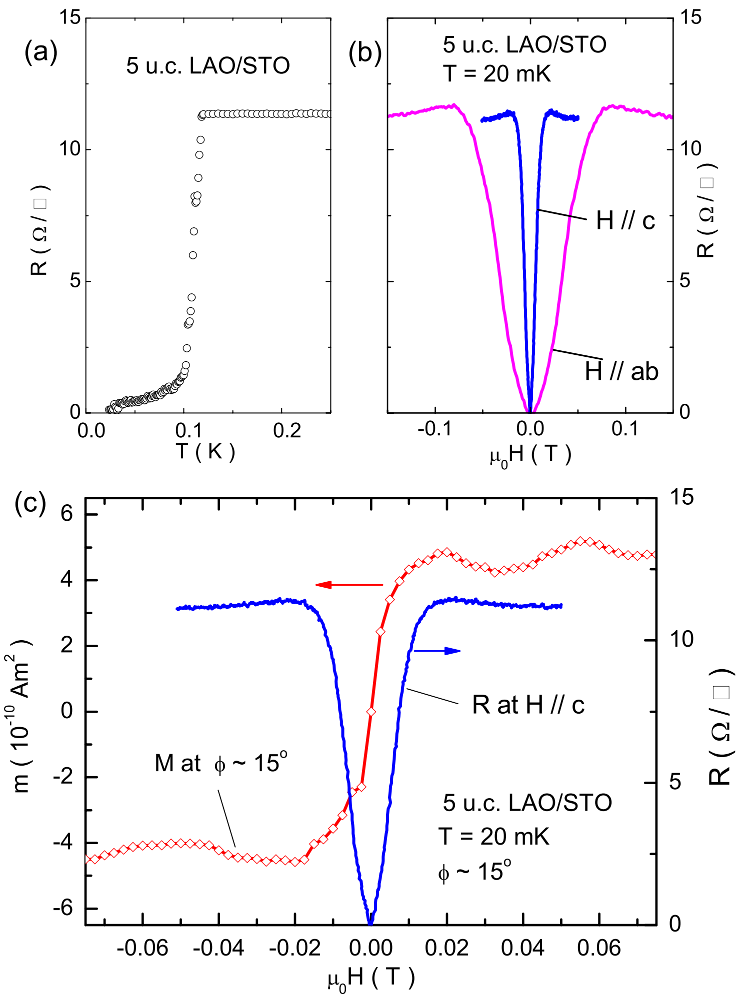

Fig. 2 **Coexistence of superconductivity and magnetic ordering in a 5 u.c. LAO/STO interface sample.** (Panel a) The temperature *T* dependence of the resistance *R* shows a superconducting transition at T_c = 120 mK. (Panel b) Field *H* dependence of *R* in different field directions taken at *T* = 20 mK. (Panel c) Field dependence of *m* measured at *T* = 20 mK at tilt angle $\phi \sim 15^{\circ}$ away from the c-axis. The $R - H$ curve is also plotted with *H* parallel to the c-axis.

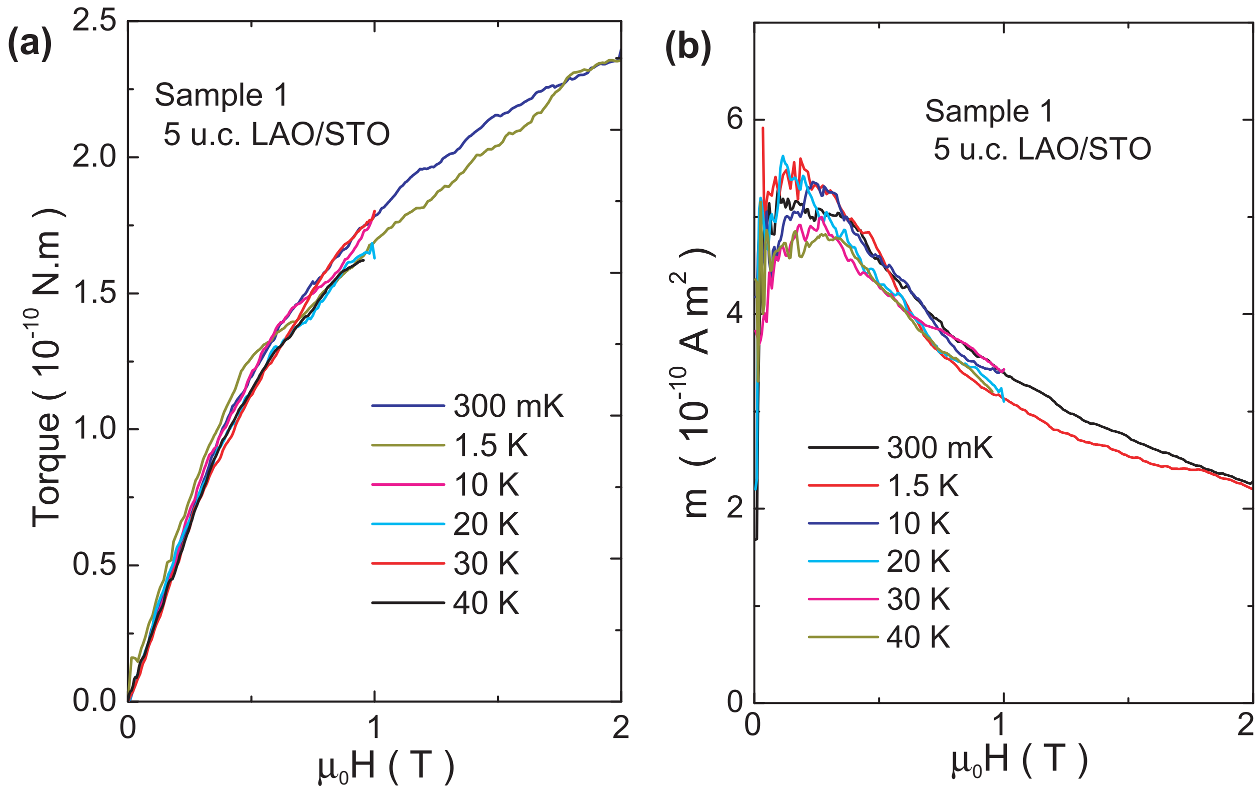

Fig. 3 **Magnetic ordering persisting to elevated temperature.** (Panel a) The torque vs. *H* curves of the 5 uc. LAO/STO sample measured at selected *T* between 300 mK and 40 K. Within the measurement noise, no strong temperature dependence is observed. The title angle is about 49°. (Panel b) The curves of *m* vs. *H* calculated from the torque curves.

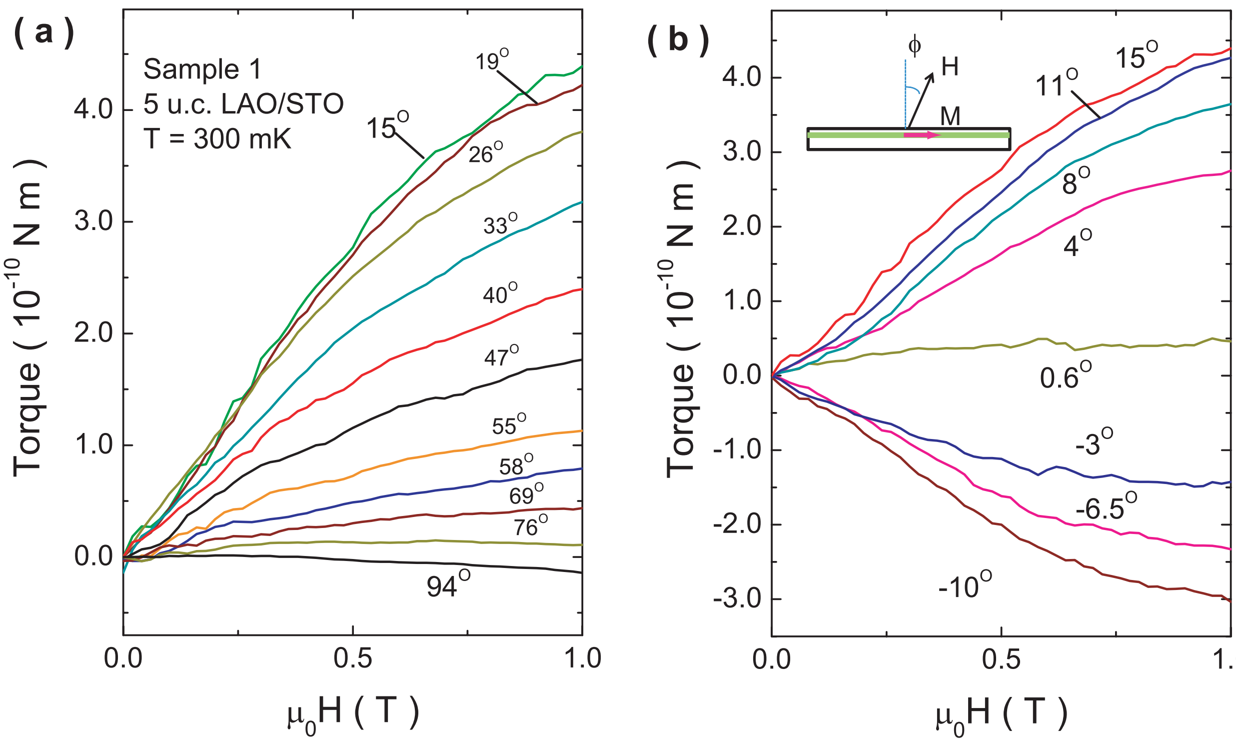

Fig. 4 **Angular dependence of the interface torque suggesting an in-plane saturation magnetic moment.** At T = 300 mK, the magnetic torque of the 5 u.c. LAO/STO sample is measured at various tilt angles ϕ between 15^o and 94^o (Panel a) and between -10^o and 15^o (Panel b). The inset of Panel b shows the geometry of the field H, magnetization M, and the definition of the tilt angle ϕ.

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