Emergent vortices at a ferromagnetic superconducting oxide interface

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Abstract
Understanding the cohabitation arrangements of ferromagnetism and superconductivity at the LaAlO3/SrTiO3 interface remains an open challenge. Probing this coexistence with sub-Kelvin magnetotransport experiments, we demonstrate that a hysteretic in-plane magnetoresistance develops below the superconducting transition for $|H_0| < 0.15$ T, independently of the carrier density or oxygen annealing. This hysteresis is argued to arise from vortex depinning within a thin (< 20 nm) superconducting layer, mediated by discrete ferromagnetic dipoles located solely above the layer. The pinning strength may be modified by varying the superconducting channel thickness via electric field-effect doping. No evidence is found for bulk magnetism or finite-momentum pairing, and we conclude that ferromagnetism is strictly confined to the interface, where it competes with superconductivity. Our work indicates that oxide interfaces are ideal candidate materials for the growth and analysis of nanoscale superconductor/ferromagnet hybrids.
Keywords: superconductor/ferromagnet hybrids, vortices, oxide interfaces

1. Introduction

The groundwork for the study of coexistent superconductivity and ferromagnetism in oxide heterostructures was laid ten years ago, when a nanoscale metallic channel was discovered at epitaxial interfaces between the band insulators LaAlO$_3$ and SrTiO$_3$ [1]. Measurements of the electrical transport in LaAlO$_3$/SrTiO$_3$ subsequently revealed a Kondo effect and hysteretic magnetoresistance (MR) [2], which are indicative of scattering off local magnetic moments and ferromagnetic ordering, respectively. This apparent emergence of ferromagnetism at the interface was a surprise, since in bulk form neither LaAlO$_3$ nor SrTiO$_3$ is magnetic, even when doped with carriers. However, $n$-type SrTiO$_3$ has been known to exhibit a superconducting ground state since the 1960s [3] and before long, superconductivity was also observed in LaAlO$_3$/SrTiO$_3$ with a critical temperature $T_c \sim 0.3$ K [4–7].

Although these early experiments reported either ferromagnetic or superconducting ground states in LaAlO$_3$/SrTiO$_3$, suggesting that the two phases might be mutually exclusive, several recent results have shown that ferromagnetism and superconductivity can indeed cohabit a single interface. A combination of torque magnetometry and resistivity data indicates a ferromagnetic moment of $\sim 0.3 \mu_B$ per interfacial unit cell coexisting with a zero resistance state [8], while scanning superconducting quantum interference device (SQUID) microscopy has imaged ferromagnetic dipoles oriented parallel to the interface, superposed on a diamagnetic (superconducting) background [9]. Perpendicular MR and Hall effect measurements on superconducting interfaces also display an unusual hysteresis below $T_c$ [10]. This apparent coexistence of ferromagnetism and superconductivity is puzzling, since ferromagnetic order breaks time reversal symmetry and is detrimental to spin-singlet superconductivity. However, spin-triplet pairing seems to be ruled out by reports of $s$-wave gap symmetry in Nb-SrTiO$_3$ [11] and LaAlO$_3$/SrTiO$_3$ [12], as well as the broken spatial inversion symmetry at the interface. It is therefore of fundamental interest to determine how ferromagnetism and superconductivity interact in LaAlO$_3$/SrTiO$_3$.

All the known coexistence mechanisms for ferromagnetism and spin-singlet superconductivity relevant to LaAlO$_3$/SrTiO$_3$ require a real-space modulation of the superconducting order parameter$^1$. This may be achieved either by spontaneous vortex formation [14], finite-momentum electron pairing, i.e. the formation of a helical Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) state due to strong Rashba spin–orbit coupling [15], or macroscopic spatial dispersion of ferromagnetism and superconductivity [9]. In the event that ferromagnetism and superconductivity are spatially-dispersed (i.e. phase-separated), an intriguing new possibility arises: the interface may function as a superconductor/ferromagnet hybrid. Hybrid materials formed from ferromagnetic layers or nanodots and superconducting films have received extensive attention [16, 17] due to their potential to act as ratchets, memory cells and spin

$^1$ We rule out any Jaccarino–Peter compensation mechanism [13] in LaAlO$_3$/SrTiO$_3$ since this heterostructure does not contain any rare earth magnetic ions and no evidence for an internal exchange field has been reported.
valves. Determining the relationship between ferromagnetic and superconducting phases at a perovskite interface and demonstrating the ability to control their interaction would constitute a major breakthrough for oxide electronics.

The development of any functional superconductor/ferromagnet capabilities must, however, be preceded by a complete understanding of the formation and location of the ferromagnetic moments in LaAlO$_3$/SrTiO$_3$; so far, this has proved elusive. Ferromagnetic behaviour in the absence of superconductivity has only been observed in the low carrier density limit, following growth and/or annealing at high O$_2$ pressures [2] to remove oxygen (O$^{2-}$) vacancies. Conversely, there is strong theoretical support for O$^{2-}$ vacancies [18, 19] or polar distortions [20] causing ferromagnetism. It is particularly important to identify whether ferromagnetism has an extrinsic (e.g. structural defects or clustered O$^{2-}$ vacancies) or intrinsic origin. Here, we may gain considerable insight by considering the spatial distribution of the magnetism, since an extrinsic origin is expected to favour inhomogeneity. Scanning SQUID microscopy has revealed an inhomogeneous distribution of ferromagnetic dipoles [9], while polarized neutron reflectometry indicates a weak magnetization (2 G) averaged across an entire macroscopic interface [21]; this also suggests that any strong emergent magnetism must be inhomogeneous. However, uncertainty still surrounds the dependence of the ferromagnetic phase on carrier density and its spatial extent; experiments have linked ferromagnetism with low [22] and high carrier densities [23], describing it as both an interfacial [24] and bulk [23] phenomenon. It is clear that a study of ferromagnetism in both stoichiometric and O$_2$-deficient interfaces over a wide carrier density range will be essential to understand not only its origin, but also its coexistence with superconductivity.

In the present work we report a hysteretic in-plane MR in LaAlO$_3$/SrTiO$_3$ heterostructures, characteristic of ferromagnetism coexisting and competing with superconductivity. This hysteresis—and hence ferromagnetic order—is always present in our samples, regardless of the carrier density or O$_2$ deficiency. We argue that the hysteresis is a consequence of vortex creation, pinning and depinning by discrete in-plane ferromagnetic dipole moments. The dipoles are tightly confined to the interface above a conventional two-dimensional (2D) superconducting layer; their confinement is confirmed by the observation of quantum oscillations from a clean, non-magnetic electron gas below the superconducting channel. No evidence emerges for a helical FFLO state; instead the principal interaction between ferromagnetic and superconducting phases is electromagnetic and long-range, i.e. vortex generation and pinning. Our magnetotransport experiments utilise the superconducting vortex dynamics as a probe for the ferromagnetic phase, thus revealing its location, origin and interaction with superconductivity. Furthermore, we may control the superconducting channel thickness and hence the vortex pinning strength using an electric field, thus providing a handle to modulate the coercive field of the ferromagnetic dipoles. Our data show that the interplay between vortices and dipoles in LaAlO$_3$/SrTiO$_3$ constitutes a uniquely tunable superconductor/ferromagnet hybrid behaviour.

### 2. Experimental details

To examine the evolution of superconductivity and ferromagnetism across the LaAlO$_3$/SrTiO$_3$ phase diagram as a function of carrier density, we have measured the ac magnetotransport in two series of heterostructures, ANN (annealed) and NAN (not annealed). All heterostructures
feature 10 unit cells (u.c.) of LaAlO\textsubscript{3} grown by pulsed laser deposition (PLD) on TiO\textsubscript{2}-terminated 0.5 mm thick SrTiO\textsubscript{3} (001) substrates, at an O\textsubscript{2} pressure of 10\textsuperscript{-3} mbar. The deposition temperature was 800 °C and the incident laser energy density 1.5 kJ m\textsuperscript{-2}. ANN-type samples were subsequently annealed for 30 minutes at 500 °C in 0.1 bar O\textsubscript{2}, in order to suppress carrier-donating O\textsuperscript{2−} vacancies: the resultant interfaces exhibit 2D carrier densities \( n_{\text{2D}} \sim 10^{13} \text{ cm}^{-2} \), which are typical of other superconducting LaAlO\textsubscript{3}/SrTiO\textsubscript{3} films studied in the literature [6, 8–10, 25]. In contrast, our NAN-type heterostructures were designed to obtain the highest possible \( n_{\text{2D}} \), i.e. beyond the maximum 3.3 \times 10^{14} \text{ cm}^{-2} imposed by the polar catastrophe. O\textsuperscript{2−} vacancy doping provides the simplest method of exceeding this limit; therefore, NAN-type samples were cooled to room temperature in 10\textsuperscript{-3} mbar O\textsubscript{2} without any high-pressure annealing, leading to interfaces with large as-grown \( n_{\text{2D}} \) in the 10\textsuperscript{14}-10\textsuperscript{15} cm\textsuperscript{-2} range (which we may further modulate by electric field-effect doping, using the SrTiO\textsubscript{3} substrate as our dielectric). All stated carrier densities are determined using Hall effect measurements (see supplementary figure S1a, available online at stacks.iop.org/njp/16/103012/mmedia).

Hall bars with Au-Ti contact pads were patterned onto the top surface of the LaAlO\textsubscript{3} using standard photolithographic techniques. We sketch this contact configuration schematically in figure 1: the obvious consequence of surface patterning is that we measure the tunneling resistance of the 10 u.c. LaAlO\textsubscript{3} in series with the interface, thus precluding the observation of zero resistance in the presence of superconductivity. However, surface contacts provide several advantages compared with directly contacting the interface, providing a qualitative estimate of the role of O\textsuperscript{2−} vacancies in our heterostructures (which will reduce the resistance of the LaAlO\textsubscript{3} layer) and simplifying the lithographic procedure. In addition, they remain sensitive to any hole-like transport at the AlO\textsubscript{2} surface resulting from the polar catastrophe doping mechanism, even when the interface is superconducting. We anticipate one further resistive component in parallel with the interface: at high carrier densities (and especially in the presence of a back-gate field \( V_g > 0 \)), it is plausible that carriers are present over a small finite depth below the interface at a
level below the critical density for superconductivity \((n_{3D} = 5.5 \times 10^{17} \text{ cm}^{-3})\) in bulk SrTiO\(_3\) [26]. At low temperature, these carriers will be 'short-circuited' by superconductivity at the interface. However, if superconductivity is quenched (either locally, due to inhomogeneity at the interface, or globally by applying a high magnetic field), these sub-interfacial states can participate in transport.

We employ a standard ac transport technique to measure the electrical resistance \(R_{xx}\) of our heterostructures, using an ac current source \((I_{ac} = 500 \text{ nA}, \nu = 19 \text{ Hz})\) and a lock-in amplifier. Experiments were carried out at temperatures down to 0.035 K in a dilution refrigerator equipped with a 9 T/4 T superconducting vector magnet, enabling us to perform a comprehensive study of the temperature and magnetic field-dependent resistance \(R_{xx}(T, H)\). The results presented in this work were acquired within a time window exceeding one year. Throughout this period our data remained entirely reproducible, thus demonstrating the long-term stability of both ferromagnetism and superconductivity at the LaAlO\(_3\)/SrTiO\(_3\) interface. Since all our samples display qualitatively similar behaviour to others within the same series, we concentrate on two specific heterostructures ANN and NAN, with as-grown \(n_{2D} = 2.3 \times 10^{13} \text{ cm}^{-2}\) and \(6.9 \times 10^{14} \text{ cm}^{-2}\) respectively. In sample NAN, an Au-Ti back-gate was sputtered across the entire base of the SrTiO\(_3\) substrate prior to LaAlO\(_3\) film growth: applying a gate voltage \(V_g = 350 \text{ V}\) increases the carrier density to \(2.4 \times 10^{15} \text{ cm}^{-2}\). Data-sets from other ANN and NAN-type heterostructures may be found in supplementary figure S2 and provide further confirmation of the reproducibility of our results.

3. Results and discussion

3.1. Kondo effect and 2D superconductivity

Evidence for inhomogeneous magnetism in our heterostructures is immediately apparent in the temperature-dependent normal-state resistance \(R_{xx}(T)\). Upon cooling from room temperature, \(R_{xx}(T)\) passes through a minimum at low temperature in both samples (figure 2(a)), then rises logarithmically prior to the onset of superconductivity at \(\sim 0.3 \text{ K}\). The minima are located at \(T_m = 10 \text{ K}\) and \(25 \text{ K}\) for samples ANN and NAN, respectively. A logarithmic divergence in the low-temperature resistance is a signature of the Kondo effect, i.e. scattering off dilute magnetic impurities\(^2\). Within the Kondo scenario, \(dR_{xx}/d \log T\) (figure 2(a) inset) is proportional to the concentration of free spins: for sample ANN, \(dR_{xx}/d \log T \sim 60 \Omega/\log K\), two orders of magnitude higher than the \(0.45 \Omega/\log K\) found in sample NAN. However, this does not necessarily imply that sample ANN contains a larger number of magnetic scattering centres; any parallel conduction from a non-magnetic electron gas will lead to a reduction in the measured Kondo resistance. Since the total number of carriers in sample NAN is more than an order of magnitude higher than in sample ANN, it is entirely plausible that a large rise in \(R(T)\) due to Kondo scattering is being masked by parallel transport through non-magnetic regions of the heterostructure. In agreement with this concept, data from a heterostructure with

\(^2\) The observed saturation in \(R_{xx}(T)\) at low temperature strongly favours a Kondo interpretation rather than weak localization, in which \(R_{xx}(T)\) should continue to diverge logarithmically as \(T \to 0\). However, we cannot completely rule out minor contributions from weak localization or Coulomb interactions to the electrical transport in our heterostructures.
n_{2D} > 10^{15} \text{ cm}^{-2} display an even smaller Kondo anomaly (figure S2), thus suggesting that the Kondo effect arises within a minority carrier population close to the interface.

The large difference in carrier density between samples ANN and NAN is also likely to be responsible for the variation in $T_m$ which we observe. $T_m$ is of the order of the Kondo temperature $T_K = \frac{1}{2} (\Gamma U)^{1/2} \exp \left[ \pi \varepsilon_0 (\varepsilon_0 + U)/\Gamma U \right]$, where $\varepsilon_0$ is the energy of the magnetic impurity relative to the Fermi level $E_F$, $\Gamma$ is the width of this impurity level and $U$ the Coulomb

\[ n_{2D} > 10^{15} \text{ cm}^{-2} \]
repulsion [28]. It should not vary significantly as the carrier density increases, and we expect $U$ to decrease due to enhanced screening. We therefore anticipate that the higher $T_K$ in sample NAN is due to a rise in $\epsilon_0$ from the increased carrier density and the corresponding rise in $E_F$. Importantly, this suggests that the same localized states are responsible for magnetism in both our heterostructures. Although $n_{2D}$ in sample ANN lies close to the Lifshitz critical density [29, 30], the Hall resistance varies linearly with magnetic field (figure S1a) and shows no signs of multiband transport. This implies that a clear majority of carriers in sample ANN occupy the lowest-lying Ti 3$d_{xy}$ band. Since the localized magnetic states must lie below the Fermi level, our data favour a $d_{xy}$ origin for ferromagnetism.

Both samples ANN and NAN are superconductors, with critical temperatures $T_c = 0.28$ K and 0.31 K, respectively. The resistive transitions in a range of magnetic fields perpendicular and parallel to the interface are shown in figure 2(b) and (c). As expected, the resistance does not vanish even at 0.035 K in zero-field, due to the series resistance from the LaAlO$_3$ layer. Assuming that the interface exhibits homogeneous superconductivity, we obtain $R$ (LaAlO$_3$) = 910 $\Omega$ and 3.4 $\Omega$ for samples ANN and NAN, thus highlighting the high O$^{2-}$ vacancy density in the LaAlO$_3$ layer of sample NAN which is responsible for its large $n_{2D}$ [31]. It should be noted that we cannot exclude inhomogeneities at the superconducting interface from contributing to our residual resistance. In fact, interfacial inhomogeneity should be expected due to cation disorder [32], the tendency of the $d_{xy}$ electrons to localize and form ferromagnetic zones [24] and dielectric inhomogeneity for $\epsilon_0 > 0$ [33]. For sufficiently high ferromagnetic zone densities close to a shallow superconducting channel, a percolative zero-resistance current path may vanish. Furthermore, in sample B it is plausible that clusters of oxygen vacancies at the AlO$_2$ surface could locally overdope the interface beyond the maximum carrier density for superconductivity; theory also suggests that such clustering will enhance ferromagnetism [18, 19, 34]. Emergent inhomogeneity is therefore a natural consequence of vacancy-doping the LaAlO$_3$/SrTiO$_3$ interface and in no way reflects negatively on our results.

We determine the thickness of the superconducting channels at our interfaces using two distinct fitting techniques to our temperature-dependent perpendicular and parallel upper critical fields $H_{c2\perp,\parallel}(T)$ (figure 2(d)). Within linearized (GL) theory [35], $H_{c2\perp}(T) = \Phi_0/2\pi\xi^2(T)$ and $H_{c2\parallel}(T) = \Phi_0\sqrt{3}/\pi\xi(T)d$, where $\Phi_0$ is the magnetic flux quantum. This yields GL coherence lengths $\xi(0) = 52 \pm 2$ nm, 60 $\pm$ 2 nm and superconducting channel widths $d = 18 \pm 1$ nm, $9 \pm 1$ nm for ANN and NAN respectively. A scaling analysis for 2D superconductors [25, 36] provides an alternative means to determine $d$ using $H_{c2\parallel}(T) = \frac{\Phi_0}{2d^2}H_{c2\perp}(T)$. This yields superconducting layer thicknesses $d = 16 \pm 1$ nm and $9 \pm 1$ nm for samples ANN and NAN, respectively, which are in excellent agreement with the values from GL theory. The larger $d$ observed in sample ANN may be caused by a partial suppression of superconductivity very close to the interface in sample NAN due to its higher carrier density. Here, it is important to remember that the as-grown vertical charge distribution profile is a key factor in determining the absolute conducting layer thickness and may vary significantly between heterostructures [7, 25, 37]. The crucial point is that $d \ll \xi$ in both ANN and NAN-type heterostructures; our samples are therefore 2D superconductors.
3.2. Phase co-existence: helical superconductivity or macroscopic dispersion?

It is of paramount importance to determine whether LaAlO₃/SrTiO₃ interfaces host a conventional superconducting phase (where we define ‘conventional’ as type-II thin film wave superconductivity). The argument for unconventional superconductivity stems from the presence of a strong Rashba spin–orbit coupling (RSOC) from broken inversion symmetry at the interface [38], which may potentially stabilize a helical FFLO state with a maximal $T_c$ for non-zero in-plane fields $H_\parallel > 0$ [15]. Since the RSOC is inversely proportional to $n_{2D}$ [7], we focus our search for exotic superconductivity on sample ANN. Figure 3(a) shows the angle-dependent upper critical field $H_{c2}(\theta)$, accurately described by the Tinkham model based on fluxoid quantization in conventional superconducting films with $d \ll \xi$ [39]:

![Figure 3](image_url)
Even at \(T = 0.1\) K, \(H_{c2}^f\) exceeds the Pauli limit \(H_p \equiv 1.84 T_c = 0.52\) T; this is consistent with the strong RSOC expected at the interface, as spin–orbit scattering is known to raise \(H_p\) [40, 41]. However, the deviations between calculated and true \(d\) are small [35] and do not impact our discussion since superconductivity remains confined within 20 nm of the interface. In figure 3(b), we track \(T_c(H_p)\) for sample ANN but find no peak at \(H_p > 0\): the data closely follow the GL model. To confirm this, we examine the MR \(R_{xx}(H_p)\) at \(T = 0.25\) K (just below \(T_c(0)\)) for both samples in figure 3(c): a maximum \(T_c\) at \(H_p > 0\) will create a point of inflection or local minimum in \(R_{xx}(H_p)\), as depicted in figure 3(d). No such features are visible, and we therefore find no support for a helical FFLO phase in our heterostructures. It should be noted that a maximum \(T_c\) for \(H_p > 0\) has been reported for LaAlO\(_3\)/SrTiO\(_3\) films grown by molecular beam epitaxy (MBE) [42], although it remains unclear whether this result is due to helical superconductivity or some other mechanism (such as quenched phase fluctuations, a field-dependent coherence length or another as-yet unidentified consequence of the strong RSOC). This difference between PLD- and MBE-grown films merits further attention, since it suggests that the 10–20 nm thick superconducting layers in PLD-grown heterostructures may be too broad for the broken symmetry of the interface to stabilize any exotic order parameter. However, \(R_{xx}(H_p)\) in our samples does reveal one unusual feature of interest: a small non-linearity at low field (leading to a peak in \(dR_{xx}/dH_p\)), which we will demonstrate to be a signature of ferromagnetism.
3.3. Hysteretic in-plane MR below the superconducting critical temperature

Figure 4 displays in-plane MR loops for both films. Hysteresis is observed at low fields \( |H| < 0.15 \) T, with peaks in \( R_{xx}(H) \) at negative/positive \( H \) after sweeping down/up from positive/negative \( H \). This pattern is distinct from the hysteresis seen in granular superconductors [43]; we may therefore immediately absolve granularity from responsibility for our data. Although hysteretic MR is generally a signature of ferromagnetism, the collapse of the hysteresis above \( T_c \) indicates that a combination of superconductivity and ferromagnetism must be responsible for our MR peaks. It should be noted that this does not imply that superconductivity is somehow responsible for the emergence of ferromagnetism. Instead, the small MR hysteresis generated by ferromagnetism in the normal state \( (T > T_c) \) falls below our experimental resolution, and we are therefore unable to determine the Curie temperature of the ferromagnetic phase.

Non-zero resistance below \( T_c \) for a superconductor in a magnetic field implies a loss of phase coherence, caused by mobile flux vortices. A vortex can be considered as a tube of diameter \( \xi \sim 2^{-1/2} \) : since our anisotropic MR indicates that \( d \ll \xi \), the superconducting channels are too shallow to allow vortex penetration parallel to an in-plane magnetic field. Instead, any vortices must be oriented within \( \pm \sin^{-1} \frac{1}{2} \) of the normal to the interface (\( < 11^\circ \) in our samples).

Interpreting our MR hysteresis therefore requires us to identify a means of generating out-of-plane vortices within the in-plane field configuration used in our experiments.

3.4. Vortex formation: the key to locating the ferromagnetic dipoles

If we assume that the applied magnetic field \( H \) lies perfectly in-plane, then the only physically viable out-of-plane flux sources in LaAlO\(_3\)/SrTiO\(_3\) are dipole fields from discrete ferromagnetic zones, polarized in-plane (figure 5(a)). To pass through the superconducting channel, the out-of-plane components of these dipole fields must be quantized. Arrays of vortices and antivortices therefore form around each pole; their size and density depend on the shape and total moment of each ferromagnetic zone. This situation is analogous to artificial 2D superconductor/ferromagnet nanodot heterostructures [16, 17].

In addition to these dipole fields, we must also consider ‘stray’ perpendicular fields, both from the Earth’s magnetic field and any misalignment of \( H \) with respect to the interfacial plane. In a bulk three-dimensional (3D) superconductor, such fields would lie far below the lower critical field \( H_{c1} = \frac{\phi_0}{2\pi d} \ln \frac{\lambda_1}{\xi} \) (where \( \lambda_1 \) is the London penetration depth) and hence be completely shielded. However, in a superconducting thin film, the screening of a field applied normal to the film is reduced since \( \lambda_1 \) is replaced by the Pearl depth \( \lambda_1 \approx \lambda d^2 = m e / \mu_0 n_s e^2 d \), where \( m \) is the electron mass and \( n_s \sim n_{2D}/d \) is the superfluid density [44]. \( \lambda_1 \gg \lambda_1 \), leading to a strong suppression of \( H_{c1} \).

As an example, consider sample ANN: using \( n_{2D} = 2.3 \times 10^{13} \) cm\(^{-2} \) and \( d = 18 \) nm we obtain \( \lambda_1 = 1.2 \times 10^{-4} \) m and \( H_{c1} = 9.6 \times 10^{-8} \) T. This represents a reduction by three orders of magnitude compared with \( H_{c1} = 3.3 \times 10^{-4} \) T for similarly doped bulk SrTiO\(_3\).

Such an exceptionally low \( H_{c1} \) enables almost any perpendicular flux source to penetrate the superconducting channel in the form of vortices. The Earth’s magnetic field strength is \( \sim 5 \times 10^{-5} \) T, corresponding to an equilibrium vortex spacing of 6.9 \( \mu \)m: since our cryostat does not incorporate any background magnetic shielding, this field alone could generate up to 2600 vortices between the voltage contacts of our Hall bars. Furthermore, we estimate a maximum
Figure 5. (a) Out-of-plane vortex creation by in-plane ferromagnetic dipoles (green rectangles). We consider three ferromagnetic zone configurations: symmetric above/below a superconducting channel (left), burial within the channel (centre), or asymmetric (right) with dipoles confined to either the top (shown) or bottom of the channel. The magnetic field due to a vortex (antivortex) points upwards (downwards). (b) Polar pinning of vortices generated by out-of-plane field components (e.g. sample misalignment or the background magnetic field from the Earth). Within our labelling convention, a small upward field component will create vortices pinned at the south poles of the ferromagnetic dipole, while a downward component forms antivortices pinned at the north poles. (c) Comparison of SdH oscillations in sample NAN upon reversing $H_y$ (data from shaded zones in figure 4(b), normalized to a polynomial background $R_{bg}(H_y)$). The oscillation amplitude and phase are field-symmetric, indicating that the electron gas below the superconducting channel is clean and non-magnetic.
error of $\pm 0.1^\circ$ in the alignment of our \textit{ab} planes with $H_\parallel$; the resultant out-of-plane flux $H_\perp = H_\parallel \sin 0.1$ exceeds $H_\perp > 5.5 \times 10^{-5}$ T. At $H_\parallel = 0.01$ T the ‘misalignment’ field $H_\parallel$ is already capable of creating a further 900 vortices in our Hall bars, and the vortex density will continue to rise linearly with $H_\parallel$. These ‘misalignment vortices’ will become pinned at the poles of discrete ferromagnetic regions [45], above or below the superconducting channel (figure 5(b)). Similar polar pinning has previously been imaged in Pb/Co nanodot hybrids [46]. As the dipole size increases, a crossover occurs from pinning misalignment vortices to independently creating vortex/antivortex pairs (figure 5(a)). Misalignment vortices are still pinned by larger dipoles, although the pole at which the vortex is pinned oscillates as the dipole moment increases [47].

We therefore have two possible sources of vortices in LaAlO$_3$/SrTiO$_3$: (a) misalignment vortices from stray perpendicular fields and (b) the out-of-plane components of in-plane ferromagnetic dipole fields. Recently, a ‘dip-hump’ MR feature was reported in W nanowires and perforated TiN films [48], which at first glance appears similar to our peaks in $R_{xx}(H_\parallel)$. This feature is caused by a giant re-entrant pinning due to vortex confinement in nanoscale device geometries. One might therefore wonder whether a similar re-entrant pinning could occur for misalignment vortices in LaAlO$_3$/SrTiO$_3$, thus removing the requirement for ferromagnetic dipoles to be present. However, a comparison of our data with this re-entrance scenario reveals a crucial difference: the MR dip-hump in [48] is permanently present, independently of the field orientation, sweep direction and magnetic history of the sample. In stark contrast, the hysteretic peak in our $R_{xx}(H_\parallel)$ data is only observed after reversing the orientation of an in-plane field. Subsequently, it does not re-appear until the field orientation is reversed once more; the absence of any peak from the Up and Down sweep data for $H_\parallel > 0$ in figure 4(b) provides an excellent example of this acute sensitivity of $R_{xx}(H_\parallel)$ to the magnetic history of the interface. This hysteresis pattern is incompatible with geometric pinning; instead, it is reminiscent of the magnetisation of a hard ferromagnet with large coercive and remanent fields. The MR hysteresis at the LaAlO$_3$/SrTiO$_3$ interface therefore corresponds to the reversal of the polarization of an array of ferromagnetic dipoles, which we will discuss in detail later in this paper.

First, we determine the location of these dipoles with respect to the superconducting channel using simple symmetry arguments. If the dipoles whose presence we have deduced are buried inside the channel (figure 5(a), centre panel) then one might expect a vortex/antivortex pair to form at each pole. However, this cannot occur due to the 2D nature of the superconducting channel: $d \ll \xi$, and so the channel is essentially homogeneous normal to the interface. The vortex–antivortex pair therefore self-annihilates, leaving a purely horizontal field within the channel. Similarly, a small in-plane dipole located inside the superconducting channel cannot pin a vortex at either of its poles, since at the south pole the attractive force from the field below the dipole is balanced by a repulsion above it (and vice versa for the north pole). Furthermore, while clearly revealing a competitive interaction, the minor destructive effect of polarized ferromagnetic zones on superconductivity (visible at $H_\parallel = 0$ in figures 4(a) and (b) as well as figure 7(b)) does not support a large dipole population inside the channel.

If the moments are instead distributed symmetrically above and below the channel (figure 5(a), left panel), then annihilation still tends to occur due to the attractive force between vortices and antivortices, leaving little quantized flux in the channel. Only an asymmetric ferromagnetic zone distribution across the channel creates stable vortex/antivortex pairs (right panel); therefore, ferromagnetism must be confined to either the top or the base of the channel.
To differentiate between these two scenarios, i.e. to determine whether the dipole moments are located above or below the superconducting channel, we directly examine the electron gas beneath this channel for any evidence of magnetism. In sample NAN, SdH oscillations develop for $H > 2.5$ T (figures 4(b) and 5(c)). Since (i) $d \leq 20$ nm, (ii) our field-effect doping proves that the SrTiO$_3$ is a good dielectric and (iii) no oscillations are visible for $H < 4$ T, there is no bulk-like 3D electron gas extending throughout the SrTiO$_3$ substrate (such as that seen in LaAlO$_3$/SrTiO$_3$ films grown at very low O$_2$ pressure [49]). Instead, our observed SdH effect is characteristic of a low carrier-density conducting ‘tail’ lying below the superconducting channel [50], penetrating the SrTiO$_3$ to a depth of at least twice the cyclotron radius $r_g = \hbar k_F/eH$ ($k_F$ is the Fermi wave vector). At $n_{2D} = 6.9 \times 10^{14}$ cm$^{-2}$, the SdH frequency $F = 25$ T; assuming Fermi surface sphericity for simplicity, $k_F = \sqrt{2\pi F/\Phi_0}$ (from the Onsager relation) and we obtain $2r_g = 88$ nm. This is much larger than the 9 nm superconducting channel at $V_g = 0$ in sample NAN; therefore, the majority of the ‘tail’ generating the SdH oscillations must lie below the superconducting channel.

SdH oscillations display clear signatures of both the scattering and magnetism within the electron gas responsible for their formation. In ferromagnetic materials, the SdH effect exhibits peak amplitude ‘beating’ and field-reversal asymmetry [51], while in dirty SrTiO$_3$ showing Kondo behaviour, scattering from localized electrons has been predicted to suppress the observation of SdH oscillations until $H \gtrsim 20$–30 T [52]. From figure 5(c), our oscillations are field-reversal symmetric, ‘beating’-free and emerge at merely 2.5 T. This implies that the electron gas below the superconducting channel is clean, non-magnetic and homogeneous: it cannot host ferromagnetic dipoles. To support this conclusion, we also consider the magnetic field distribution from any dipoles lying below the superconducting channel: the vertical component of the dipole field will be quantized as it passes through the channel, strongly distorting the flux profile when viewed from above. No evidence for any such distortion has been seen by scanning SQUID microscopy [9, 53, 54]. We deduce that our SrTiO$_3$ substrates are non-magnetic: the ferromagnetic dipoles must therefore be confined to the interface, above the superconducting channel. Here, they create and/or pin vortices as depicted in figure 5(a) (right panel) and 5(b).

We briefly consider the other possible sources of vortices in LaAlO$_3$/SrTiO$_3$ which have previously been discussed in the literature. Spatial inhomogeneities in the RSOC or in-plane polarization can in principle create vortices in helical superconductors [38], but—apart from the fact that we find no evidence for helical superconductivity—these only emerge above a critical field and are hence incompatible with our low-field hysteresis. It has been suggested that Bloch domain walls in a continuous ferromagnetic layer may generate vortices, which then propagate when the magnetic field is swept [55]. However, this scenario is unlikely for several reasons. Firstly, shape anisotropy is a key factor in domain formation for thin ferromagnetic layers, and the walls should be of Néel rather than Bloch-type, i.e. their magnetization points in-plane and the out-of-plane flux is greatly reduced. A recent magnetic force microscopy study of carrier-depleted LaAlO$_3$/SrTiO$_3$ has indeed confirmed the presence of Néel-type walls [56]. Secondly, there is no evidence in the literature for a continuous ferromagnetic layer coexisting with superconductivity; in contrast, SQUID microscopy [9] and theoretical considerations [34] both indicate inhomogeneous ferromagnetism. Thirdly, we acquire $R_{xx}(H)$ data in stable fields with our superconducting magnet in persistent mode, i.e. $dH/dt = 0$, so there is no driving force for domain-wall propagation (and our ac current negates spin–torque transfer effects on the
Finally, equilibrium domain formation in a ferromagnet is driven by the requirement to minimize the total free energy by balancing the exchange, anisotropy, magnetostatic and Zeeman energies. The domain structure therefore evolves with the applied field, which should generate mobile vortices (and hence a rise in the MR) regardless of the field sweep direction. This pattern is not present in our data: no peak or anomaly in $R_{xx}(H_{||}H_{0})$ is observed upon sweeping large in-plane fields down to zero. We therefore find no evidence for mobile domain walls in our LaAlO$_3$/SrTiO$_3$ heterostructures.

**Figure 6.** (a) Ferromagnetic dipole repolarization upon sweeping from $H_{||} \gg 0$ to $H_{||} \ll 0$. Green and purple denote opposing polarizations; vortices are red and antivortices blue. Yellow arrows illustrate dipole rotation for $|H_{||}| > H_{c0,0}$, dragging vortex/antivortex pairs with them. (b) Amplitude variation of the hysteretic peak in $R_{xx}(H_{||})/R_{xx}(0T)$ for three time-dependent field sweeps (see text for details; data from an ANN-type heterostructure). Sweeps were performed after polarization at $H = -4$ T. Each data-point was acquired in a stable, constant field, 90 s after our superconducting magnet entered persistent mode; the ac measurement current flowed throughout the experiment. The field increment between data-points determines the measurement duration (inset) and hence the peak amplitude. The peak shape (related to the channel pinning profile) is stable between sweeps, but exhibits small changes after thermal cycling to 300 K.
3.5. Vortex motion during ferromagnetic polarization reversal

Having confirmed that a series of discrete ferromagnetic dipoles above the superconducting channel is responsible for vortex formation in LaAlO₃/SrTiO₃, we now consider the physical mechanism driving the hysteretic MR in detail. Peak formation in $R_{xx}(H)$ arises due to ferromagnetic polarity reversal, a process illustrated in figure 6(a). This mainly occurs via dipole rotation rather than domain wall propagation due to the limited zone size ($\lesssim 10\,\mu\text{m}$ from SQUID data), the presence of hysteresis up to $|H| = 0.15$ T (implying a large coercive field $H_{\text{coerc}}$) and flux conservation within the channel. During repolarization, a torque $\mathbf{m} \times \mathbf{H}$ acts on ferromagnetic zones, where $\mathbf{m}$ is the dipole moment. The vortex/antivortex arrays around the poles are rotated through 180° in-plane: while mobile, the vortices develop an electric field in their cores and dissipate energy, thus creating a hysteretic peak in $R_{xx}(H)$. Dipoles which are too small to independently generate vortex/antivortex pairs contribute to the observed hysteresis by pinning misalignment vortices at their poles; these vortices are also rotated around their respective dipoles upon ferromagnetic polarity reversal. However, all vortex motion is impeded by pinning due to impurities within the underlying superconducting channel, which broadens the MR peak and increases the effective coercive field of the dipoles. Within the framework of the Anderson flux creep model [57], we may write:

$$\nu_{\text{depin}} \propto e^{-(U - |m|H + m\mathbf{H})/k_BT},$$

where $\nu_{\text{depin}}$ is the depinning frequency, $U$ is the pinning potential, $|H| > H_{\text{coerc}}$, and we disregard depinning by the ‘shaking’ effects of our ac current. $\nu_{\text{depin}}$ scales linearly with the induced electric field and hence $R_{xx}(H)$. Assuming an infinite vortex supply, constant $U$ and ferromagnetic polarization antiparallel to $H_g$, an increase $\Delta H$ will generate $\Delta R_{xx}(H_g) \propto e^{2mH\Delta H}$. Such assumptions are unrealistic, since our samples contain finite numbers of vortex-inducing ferromagnetic dipoles which may be pinned at arbitrary angles to $H_g$ during repolarization; we also expect local variation in the superconducting channel pinning potential from inhomogeneous defect distributions. Nevertheless, this relation permits a qualitative understanding of the temporal evolution of our hysteretic peaks (figure 6(b)). Compared with a reference data-set (sweep 1) acquired at field increment $\delta H = 0.005$ T, a long pause in the data acquisition at $H_{g} = 0.005$ T (sweep 2) has little effect on $R_{xx}(H_g)$, since $H_{\text{coerc}} > 0.005$ T for most of the dipoles and depinning is limited. In contrast, measuring with no pause but a smaller increment $\delta H/3$ (sweep 3) yields a larger drop in $R_{xx}(H_g)$, since fewer vortices are depinned at each data point. This dependence of $R_{xx}(H_g)$ on the measurement duration cannot be explained within a geometric pinning scenario [48]. Above $H_g \sim 0.15$ T, ferromagnetic repolarization is complete: $R_{xx}(H_g)$ therefore falls to its background level.

For completeness, we also consider the MR in a field perpendicular to the interface. Small fields $H_L < H_{c2\perp}$ will create a high density of vortices pinned at the poles of the ferromagnetic dipoles; however, reversing the direction of $H_L$ does not change the in-plane orientation of the dipoles. Increasing the field, a combination of large magnetic anisotropy and strong vortex pinning implies that the dipoles cannot be rotated out-of-plane for $H_L < H_{c2\perp} \sim 0.1$ T. Therefore, no dipole-induced vortex motion can occur in perpendicular fields, and hence no large hysteretic peaks of the type seen in $R_{xx}(H_g)$ are expected for $H_L$. In agreement with this prediction, no hysteretic peaks are indeed visible in our measured $R_{xx}(H_L)$ (figure S1b), although a very small hysteresis develops at fields close to zero, comparable to the typical remanence in superconducting magnets. The absence of any reversible (i.e. independent of the
field sweep direction) dip-hump feature in our $R_{xx}(H_{\perp})$ data confirms that geometric pinning does not play a significant role in the vortex dynamics of our heterostructures [48].3 Furthermore, the lack of any peaks in $R_{xx}(H_{\perp}| 0)$ is entirely consistent with our conclusion that the hysteretic peaks in $R_{xx}(H_{\perp})$ below $T_c$ are primarily a consequence of the interaction between vortices and in-plane ferromagnetic dipoles.

3.6. Electric field tuning of the vortex pinning

Applying a back-gate voltage $V_g > 0$ to a LaAlO$_3$/SrTiO$_3$ heterostructure (see figure 1(a)) should increase the superconducting channel thickness $d$, since electrons are simultaneously injected at the interface and ‘decompressed’ into the SrTiO$_3$ substrate. We confirm this quantitatively in figure 7(a), where we plot $H_{c2\perp}$ for sample NAN at $V_g = 350$ V. G-L and scaling analyses both indicate that $d$ is approximately doubled from its $V_g = 0$ V value of 9 nm.

SQUID microscopy studies have indicated that gating the LaAlO$_3$/SrTiO$_3$ interface to modulate its carrier density does not have an effect on the density of ferromagnetic inclusions [53, 54]. This suggests that the total number of vortices within the superconducting channel is unlikely to change with $n_{2D}$. However, the Pearl magnetic penetration depth $\lambda_{\perp}$ will fall as $n_{2D}$ increases, leading to a decreased pinning efficiency. Our time-dependent data (figure 6(b)) show that depinning controls the hysteresis in $R_{xx}(H_g)$, so we anticipate some variation in the shape of the hysteretic peaks upon changing $V_g$. This is indeed observed: the peaks at $V_g = 350$ V are broader and begin to form at smaller $H_{\perp}$ than at 0 V (figure 7(b)).

We attribute the onset of hysteresis at lower fields to weaker pinning due to a reduction in $\lambda_{\perp}$, since $n_{2D}$ has risen from $6.9 \times 10^{14}$ to $2.4 \times 10^{15}$ cm$^{-2}$ (figure S1a). The fall in $\lambda_{\perp}$ is partially offset by the rise in $d$, which increases the number of available pinning centres in the superconducting channel. In turn, this augments the probability of pinning any given vortex

3 It should be noted that the lateral dimensions (e.g. nanowire width, perforation period) of SC channels that exhibit re-entrant pinning are typically $\leq 10^{2}$. In contrast, the width of our Hall bars is 80 $\mu$m, i.e. $> 1300^{2}$. Even allowing for some inhomogeneity in our SC channel, it seems unlikely that geometric confinement could play an important role in the vortex dynamics of our heterostructures.
during the rotation of its respective ferromagnetic dipole, which will broaden the hysteretic peaks. However, the strongest pinning is likely to originate from isolated structural defects close to the interface and is thus independent of $\lambda_\perp$ and $d$. The maximum field at which hysteresis is observed should therefore remain similar regardless of the applied $V_g$, which is in agreement with our data.

4. Conclusions

The overall picture emerging from our work is of a narrow layer of discrete ferromagnetic dipoles, confined above a 2D superconducting channel at the LaAlO$_3$/SrTiO$_3$ interface. These dipoles are capable of generating vortices within the superconducting channel, as well as pinning vortices created by weak extrinsic out-of-plane fields.

Our data also clarify much of the confusion that has surrounded the origin of ferromagnetism in LaAlO$_3$/SrTiO$_3$. The inhomogeneity in the ferromagnetism which we deduce from our data and its independence from $n_{2D}$ suggest that it originates from $d_{xy}$ electrons localized by static defects, in agreement with recent models [18–20]. Previous links between ferromagnetism and high O$_2$ pressure growth [2, 58] arise from enhanced defect-induced localization in the 2D limit and ferromagnetic transport signatures being ‘short-circuited’ by mobile electrons below the interface at higher $n_{2D}$. Together with data from such resistive interfaces, our work shows that ferromagnetism can exist across the entire LaAlO$_3$/SrTiO$_3$ phase diagram: $10^{12} \leq n_{2D} \leq 10^{15}$ cm$^{-2}$. The similarities in the hysteretic MR of our two films indicate that O$^{2−}$ vacancies are neither essential nor anathemic to ferromagnetism; however, scattering from vacancies and cation intermixing may help to stabilize ferromagnetism. The Kondo effect and the ferromagnetism which we infer from our hysteretic MR are both consequences of defects, but at different length scales: isolated localized electrons at the interface constitute the magnetic scattering centres that are responsible for the Kondo effect, while larger groups of localized electrons (formed by O$^{2−}$ vacancy clustering, for example) order ferromagnetically to create in-plane dipoles. It remains an open question whether this ordering is mediated by direct exchange, double exchange or the RKKY interaction, and we hope that our results will stimulate more work in this field. We also note that beyond the hysteretic in-plane MR and Kondo effect in $R_{xx}(T)$, there is no obvious evidence for magnetism in our heterostructures. It is likely that ferromagnetism in other LaAlO$_3$/SrTiO$_3$ films studied in the literature was overlooked, since parallel MR loops were not acquired below $T_c$.

More generally, our work illustrates how reduced symmetry, modified electronic structure and defects at an interface enable ferromagnetism—an emergent phase alien to both LaAlO$_3$ and SrTiO$_3$ in bulk form—to interact and compete with the usual ground state in doped SrTiO$_3$ (superconductivity). Although competition only occurs at the top of the superconducting channel, the asymmetric distribution of ferromagnetism influences the entire superconducting layer by generating vortices even at zero applied field. The ability to continuously tune the vortex pinning by varying the superconducting channel thickness (and hence influence the dipole coercive field) represents a major advantage of oxide interfaces over any other known superconductor/ferromagnet hybrids. In the future, we suggest that atomically precise defect control may enable the development of advanced hybrid superconductor/ferromagnet devices in LaAlO$_3$/SrTiO$_3$, such as spintronic latches, vortex memory or superconducting qubits.
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