Tuning spin-orbit coupling and superconductivity at the SrTiO$_3$/LaAlO$_3$ interface: a magneto-transport study

M. Ben Shalom, M. Sachs, D. Rakhmilevitch, A. Palevski, and Y. Dagan$^\dagger$

$^\dagger$Raymond and Beverly Sackler School of Physics and Astronomy, Tel-Aviv University, Tel Aviv, 69978, Israel

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The superconducting transition temperature, $T_c$, of the SrTiO$_3$/LaAlO$_3$ interface was varied by the electric field effect. The anisotropy of the upper critical field and the normal state magneto-transport were studied as a function of gate voltage. The spin-orbit coupling energy $\epsilon_{SO}$ is extracted. This tunable energy scale is used to explain the strong gate dependence of the mobility and of the anomalous Hall signal observed. $\epsilon_{SO}$ follows $T_c$ for the electric field range under study.

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Interfaces between strongly correlated oxides exhibit a variety of physical phenomena and are currently at the focus of intensive scientific research. An electronic reconstruction occurring at the interfaces may be at the origin of these phenomena.\cite{1} It has been demonstrated that the interface between SrTiO$_3$ (STO) and LaAlO$_3$ (LAO) is highly conducting, having the properties of a two dimensional electron (2DEG) gas.\cite{2} At low temperatures the 2DEG has a superconducting ground state, whose critical temperature can be modified by an electric field effect.\cite{3} The origin of the charge carriers and the thickness of the conducting layers are still under debate.\cite{3,4–6} Recently, we have demonstrated that for carrier concentrations at the range of $10^{13}$ cm$^{-2}$, a large, highly anisotropic magnetoresistance (MR) is observed.\cite{10} Its strong anisotropy suggests that it stems from a strong magnetic scattering confined to the interface.

Here we show that both superconducting and spin-orbit (SO) interaction can be modified by applying a gate voltage. The upper critical field applied parallel to the interface, $H_{c2}||$, is approximately the weak coupling Clogston-Chandrasekhar paramagnetic limit: $\mu_0 H_{c2} \approx 1.75 k_B T_c$ for high carrier concentrations. However, as the density of the charge carrier is reduced, $H_{c2}||$ becomes as large as five times this limit, suggesting a rapidly increasing SO coupling. This SO coupling energy $\epsilon_{SO}$ manifests itself in many of the transport properties studied.

We use a sample with 15 unit cells of LaAlO$_3$, deposited by pulsed laser on an atomically flat SrTiO$_3$(100) substrate. The oxygen pressure during the deposition was maintained at $10^{-4}$ Torr. The deposition was followed by a two-hour annealing stage at oxygen pressure of 0.2 Torr and a temperature of 400 C. The thickness of the LaAlO$_3$ layer was monitored by reflection high energy electron diffraction. Growth procedure is similar to the one described elsewhere.\cite{10} A layer of gold was evaporated at the bottom of the sample and used as a gate when biased to $\pm 50$ V relative to the 2DEG. Contacts were made using a wire bonder in a Van Der Pauw geometry. We took extra care to ensure the absence of heating by testing the resistance to be current-independent at the superconducting transition point, where current sensitivity is maximal. During measurements, the sample was cooled down to a base temperature of 20 mK and a magnetic field of up to 18 T was applied. Samples rotation was done using a step motor with a resolution of 0.015$^\circ$/step. All transport properties reported here were measured in the as-grown state (AG), and while applying $V_g=50$, 10, -10, -50 V. In order to accurately determine $T_c$, the remnant magnetic field was minimized by oscillating the field down to zero. In addition, the sample was kept parallel to the field where superconductivity is less sensitive to it.

Fig 1a presents the normalized resistance $R/R_n$ as a function of temperature for the various $V_g$ values. $R_n$ is the resistance measured at zero field and at $T=0.5$ K, well above $T_c$, for each $V_g$. Tuning $V_g$ from $+50$ to $-50$ V increases $T_c$ from 0.1 to 0.35 K.

Fig 1b presents the resistance normalized with $R_n$ versus the perpendicular magnetic field at $T=20$ mK.
FIG. 2: Sheet resistance versus the magnetic field applied parallel to the interface and current for various charge densities.

A large, positive MR is observed for \( V_g = 50 \) V. Its magnitude gradually decreases as \( V_g \) is reduced. The inset zooms on the superconducting low field region.

Table 1 summaries the superconducting properties for the various \( V_g \). We define \( H_{c2 \perp} \) (\( T_c \)) as the field (temperature) at which the resistance is \( R_n/2 \). The Ginzburg Landau coherence length \( \xi_{GL} = \sqrt{\Phi_0/(2\pi\mu_0 H_{c1 \perp})} \) is extracted from \( H_{c2 \perp} \), here \( \Phi_0 \) is the flux quantum.

The charge carrier density \( n = 1/RHc \) is inferred from Hall measurements in the high field regime (\( \mu_0 H > 14 \) T). It is presented together with the calculated mobility \( \mu = n/R_n \). A large variation of \( \mu \) is observed, indicating a significant change in the scattering rate with gate voltage as previously noted. This behavior will be discussed later.

Fig. 2 presents the sheet resistance versus the magnetic field, \( H_{\|} \), applied parallel to the interface and current. From the low field regime we extract the superconducting parallel critical field \( H_{c2 \|} = H(R_n/2) \). We note that while \( T_c \) increases merely to 0.35 K, \( H_{c2 \|} \) becomes as large as 2.5 T [table 1]. The high \( H_{c2 \|} \) and low \( T_c \) implies that the paramagnetic limit is exceeded. [12] Since \( \xi_{GL} \) is rather large, 50-200 nm, it is reasonable to use the weak coupling BCS approximation. Accordingly, we expect superconductivity to exist in fields lower than \( g\mu_B H \leq 3.5 k_B T_c \); here \( g \) is the gyromagnetic ratio and \( \mu_B \) is the Bohr magneton.

In Fig. 3, \( \mu_B H_{c2 \|} \) is plotted against \( k_B T_c \) for different \( V_g \) values. The dashed straight line represents the paramagnetic limit using BCS weak coupling and \( g = 2 \). As \( T_c \) increases, the limit is exceeded by up to a factor of \( \sim 5 \). This behavior suggests a strong SO coupling that relaxes the Clogston-Chandrasekhar limitations. For a superconducting layer thickness \( d \ll \xi_{GL} \) it is expected that \( H_{c2 \|} \) be much larger than \( H_{c2 \perp} \). In this case the critical field is determined by the paramagnetic limit and the spin-orbit energy. From Fig. 3, it appears that even for the lowest \( T_c \), superconductivity is quenched in this paramagnetic limit. We can therefore merely set an upper limit on the thickness of the conducting layer by analyzing the anisotropy of the critical field: \( d \leq \sqrt{3}\Phi_0/\pi \xi_{GL} \mu_0 H_{c2 \perp}. \)\[13\] The thickness upper limit is presented for the various \( V_g \) reaching a minimal value of 10 nm for \( n = 3 \times 10^{13} \) cm\(^{-2} \) [table 1]. Previous estimations obtained similar values. They, however, relate \( d \) to the actual thickness.\[11\][12]

Let us describe the general behavior of \( R(H_{||}) \) [Fig. 2] using the \( n = 6.1 \times 10^{13} \) cm\(^{-2} \) curve (green triangle) as an example: Above the superconducting transition, the sheet resistance reaches a roughly field-independent regime (0.9-1.8 T) in which the resistance approximately equals \( R_n \). We define \( H^* \) as the onset field where \( dR/dH \) becomes negative. For \( H > H^* \) the resistance drops to \( R_{sat} \), the low resistance saturation regime (\( H_{||} > 10 \) T).

Both \( H_{c2 \|} \) and \( H^* \) strongly depend on \( V_g \); they increase as \( V_g \) changes from 50 to -50 V, and \( H^* \) becomes unmeasurably high for \( V_g = -50 \) V (black squares).

| \( V_g \) | \( n \times 10^{13} \) | Mobility | \( T_c \) | \( \mu_B H_{c1 \perp} \) | \( \mu_B H_{c2 \perp} \) | \( d \) | \( \xi_{GL} \) | \( \epsilon_{SO} \) |
|------|----------------|---------|------|----------------|----------------|------|---------|---------|
| -50  | 3.0            | 236     | 0.35 | 0.125         | 2.23           | 10   | 51      | >2      |
| -10  | 4.4            | 392     | 0.28 | 0.098         | 1.37           | 14.4 | 58      | 0.58    |
| 10   | 6.1            | 762     | 0.18 | 0.040         | 0.52           | 24.1 | 90      | 0.21    |
| 50   | 7.8            | 1707    | 0.12 | 0.009         | 0.14           | 41.2 | 194     | 0.06    |
| AG   | 9.5            | 897     | 0.15 | 0.030         | 0.25           | 44   | 103     | 0.12    |

Fig. 3 suggests that SO coupling plays a major role in the system. We shall now describe the data presented so far using a single energy scale, \( \epsilon_{SO} \). We relate \( H^* \) to the breakdown field of the spin-orbit coupling energy \( g\mu_B H^* = \epsilon_{SO} \). The \( \epsilon_{SO} \) values are presented assuming \( g = 2 \) [table 1]. Above this field SO scattering is suppressed and completely vanishes at the high field saturation regime where all spins are aligned. In this scenario the high field saturation value \( R_{sat} \) is the remnant impurity scattering. The SO scattering rate can be evaluated from the difference between \( R_n \) and \( R_{sat} \). Therefore \( R_n - R_{sat} \) should be proportional to \( h/\tau_{SO} = \epsilon_{SO} \).

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transport and affects superconductivity. For the carrier concentration range under study $R_{\text{sat}} \ll R_n$, hence, the main contribution to the zero field resistance comes from SO scattering. This explains the strong dependence of the mobility on gate voltage.

We note that for $V_g = 50$ V the carrier concentration is lower compared to the as-grown state. This is in contrast to a simple capacitor-like behavior with negative charge carriers. Furthermore, despite the decrease in carrier concentration, $R_n$ unexpectedly decreases. We relate this peculiar behavior to the SO scattering processes dominating $R_n$. We assume that static positive charges move to the interface when the highest positive voltage is applied and consequently $n$ is reduced. This charge movement reduces the initial electric field at the interface and as a result decreases the SO scattering. Upon reducing the gate voltage from its maximal positive value, electrons are drawn away from the interface in a reversible manner as expected, and an electric field is created. The initial electric field in the as-grown state results in enhanced SO scattering and higher resistance, despite the higher carrier concentration.

The MR presented [Fig.1b, Fig.2] is highly anisotropic for high magnetic fields. For example, for $n = 6.1 \times 10^{13} \text{cm}^{-2}$ and $H=18$ T, the resistance vary from 0.12$R_n$ to 3.5$R_n$ (a factor of 30) for the in-plane and out-of-plane field configurations respectively. We study this anisotropy by rotating the sample around an in-plane axis which is perpendicular to the current, while keeping the total field constant, $|\vec{H}| =18$ T. Fig.1b presents the normalized sheet resistance versus the perpendicular field component. For the small angle range presented, the parallel field component is approximately constant $\approx 18$ T. Moreover, for this parallel field component, the resistance is insensitive to small changes in $H_\|$ [Fig.2]. As shown, the sheet resistance rapidly increases when a small perpendicular component is introduced. It reaches $R_n$ for $H_\perp \approx 1.5$ T (an angle of about 4°). This is similar to our previous observation at 2 K. The curves $n = 7.8, 6.1 \times 10^{13} \text{cm}^{-2}$ and AG (not shown) merge near 1.5 T, while $n = 4.4 \times 10^{13} \text{cm}^{-2}$ departs form the general behavior. This can be easily explained since for this gate voltage the resistance does not saturate up to 18 T [Fig.2]. For $n = 3.0 \times 10^{13} \text{cm}^{-2}$, $H=18$ T is well below $H^*$ and therefore the anisotropy cannot be observed at this too small a magnetic field. We relate the sudden appearance of resistance with a small perpendicular field component to the orbital motion generated by this component. This motion is enhanced by the high mobility state ($R_\perp R_{\text{sat}}$) induced by the presence of a large parallel magnetic field.

Fig.2b presents the Hall resistivity $\rho_{xy}$ at 20 mK versus the magnetic field after subtracting out the linear term obtained from a fit to the high field regime. A conspicuous deviation from this linear part in the Hall resistivity (anomalous Hall effect AHE), is observed. This AHE persists up to 100 K with a peculiar, roughly linear temperature dependence (not shown). As shown, the applied $V_g$ varies the AHE saturation field. This variation is inconsistent with a simple magnetic impurity scenario and with a simple ferromagnetic behavior. Furthermore, no hysteresis is observed for all carrier concentrations. Here we note that the AHE saturation field roughly follows the behavior of $\epsilon_{SO}$ and may be related to it.

FIG. 3: (Color on-line) a. The parallel upper critical field $H_{c1}$ versus $k_BT_c$ while $V_g$ is varied. The dashed line represents the expected behavior for the paramagnetic limit. b. $\epsilon_{SO}$ extracted from $H^*$ [see Fig.2], and calculated from the measured superconducting properties ($H_{c1}$ and $T_c$) [17] versus $k_BT_c$. The right axes presents $\Delta R$, which scales with $\epsilon_{SO}$ as expected.

Spin-orbit interactions are expected to play a signifi-
significant role at the interface due to the inversion symmetry breaking. The SO interaction term is of the form: 
\[ \epsilon_{SO} = p \cdot \sigma \times \nabla V \] 
where \( p \) is the carrier momentum, \( \sigma \) is the Pauli spinor, and \( \nabla V \) is the electric field perpendicular to the interface in our system.\[10\] The strong gate dependence of \( \epsilon_{SO} \) directly follows from this expression. As pointed out above, the initial local electric field is unknown. When a positive gate voltage was applied for the first time the number of charge carriers decreased, while the resistivity decreased. We believe that this is mainly due to a decrease in the total electric field near the interface, resulting in a decreased SO scattering and an enhanced mobility. This suggests that the initial field (AG state) is a consequence of the electronic reconstruction. In this scenario, adding more LaAlO\(_3\) layers increases the as-grown field and hence the SO scattering. This explains the decrease in mobility while increasing the LaAlO\(_3\) thickness.\[20\]

It seems that most scattering processes in the normal-state involve a spin flip, which strongly impede the transport. In a simple metal such processes do not contribute much to the resistivity, yet in our system they become important due to the strong SO coupling. This is clear in the case of in-plane spin flip processes; since SO coupling affects in-plane spin orientations, a spin flip process results in momentum reversal and consequently in a strong contribution to the resistivity. These spins are strongly coupled to the in-plane momenta; therefore it is impossible to align them with \( H_3 \) unless the energy associated with the field \( g\mu_B H_3 \) exceeds \( \epsilon_{SO} \). For \( H>H^* \), the spins gradually align along the field direction and so \( R_{in} \) is reduced. When all spins are aligned, the resistance reaches the saturation value \( R_{sat} \). For the out-of-plane field orientation, suppression of spin scattering is overwhelmed by the large positive MR.

Tuning the gate voltage from 50 to -50 V increases the local electric field, resulting in an increase of \( \epsilon_{SO} \) and \( H^* \). Unlike \( R_{in}\) (the zero field resistance), \( R_{sat}\) roughly scales with number of charge carriers deduced from the high field Hall measurements. This suggests that \( R_{sat}\) is a result of standard impurity scattering. For \( V_g=-50\) V, \( \epsilon_{SO} \) is extremely large and an in-plane field of 18 T is not enough to align the spins and suppress the spin-scattering resistivity. Upon applying a small perpendicular field component and due to the large scattering time, orbital motion is immediately turned on along with the SO coupling. This results in a full recovery of the spin-MR.

In summary, we study the phase diagram of the SrTiO\(_3\)/LaAlO\(_3\) interface in the region where \( T_c \) increases, while reducing the carrier concentration by variation of gate voltage. We demonstrate the important effect of spin-orbit (SO) interaction on both superconductivity and on normal-state transport. The SO coupling energy (\( \epsilon_{SO} \)) is evaluated using two independent transport properties, and is also in agreement with theoretical model given the superconducting parameters: \( T_c \) and the upper critical parallel field. \( \epsilon_{SO} \) follows \( T_c \) for the electric field range studied. Our results suggest that SrTiO\(_3\)/LaAlO\(_3\) interfaces may be useful for future oxide based devices controlling the orbital motion of electrons by acting on their spins.\[21\]

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* yodagan@post.tau.ac.il

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