Spin-orbit-coupled superconductivity

Shun-Tsung Lo1, Shih-Wei Lin2, Yi-Ting Wang3, Sheng-Di Lin2 & C.-T. Liang1,3

1Graduate Institute of Applied Physics, National Taiwan University, Taipei 106, Taiwan, 2Department of Electronics Engineering, National Chiao Tung University, Hsinchu 300, Taiwan, 3Department of Physics, National Taiwan University, Taipei 106, Taiwan.

Superconductivity and spin-orbit (SO) interaction have been two separate emerging fields until very recently that the correlation between them seemed to be observed. However, previous experiments concerning SO coupling are performed far beyond the superconducting state and thus a direct demonstration of how SO coupling affects superconductivity remains elusive. Here we investigate the SO coupling in the critical region of superconducting transition on Al nanofilms, in which the strength of disorder and spin relaxation by SO coupling are changed by varying the film thickness. At temperatures $T$ sufficiently above the superconducting critical temperature $T_c$, clear signature of SO coupling reveals itself in showing a magneto-resistivity peak. When $T < T_c$, the resistivity peak can still be observed; however, its line-shape is now affected by the onset of the quasi two-dimensional superconductivity. By studying such magneto-resistivity peaks under different strength of spin relaxation, we highlight the important effects of SO interaction on superconductivity.

Spin-orbit (SO) interaction, which couples the electron orbital motion to its spin, has been at the center of intensive research efforts in the field of spintronics1–3. This coupling between orbital motion and spin orientation leads to several interesting physical phenomena such as the spin Hall effect4,5, spin-orbit gap6,7 and spin relaxation8,9, and may open an avenue to achieve all-electrical control of spin degree of freedom10. In addition, SO interaction provides an essential ingredient in creating topological insulators11 and Majorana fermions12. Furthermore, recently SO coupling was found to have a significant impact on quasi two-dimensional (2D) superconductivity. One of the most fascinating examples is the promotion of superconductivity with enhanced strength of SO interaction13. In the 2D electron gas at the interface between LaAlO3 and SrTiO3 exhibiting superconducting properties at low temperatures $T$, large Rashba SO coupling arising from interfacial breaking of inversion symmetry is shown to have pronounced effects in stabilizing the superconducting state13,14. Moreover, due to strong SO coupling at the interface, ferromagnetism, which is usually thought to be incompatible with conventional superconductivity, can now coexist with superconductivity15,16. Interestingly, SO coupling also appears relevant to the observed increase of superconducting critical temperature $T_c$ by an in-plane magnetic field in Pb thin films17. Within the framework of the Bardeen-Cooper-Schrieffer (BCS) theory18 which attributes superconductivity to pairs of electrons with opposite spin projections (Cooper pairs), any mechanism that causes spin alignment (like those induced by paramagnetic impurities and magnetic fields) is not predicted to enhance superconductivity. Response of superconductivity to magnetic perturbations can be fundamentally different in the presence of SO coupling.

In a quasi 2D superconductor with SO coupling, the application of a magnetic field perpendicular to the plane of carrier motion may not only lead to suppression of weak antilocalization (WAL) induced by SO coupling19–24, but also to orbit de-pairing of Cooper pairs, destroying the zero-resistivity superconducting state. Magneto-transport studies could thereby allow one to obtain fundamental information regarding the interplay between SO coupling and superconductivity. Here we present such measurements on molecular-beam epitaxy (MBE)-grown Al nanofilms with thicknesses of 3 nm, 6 nm, and 12 nm in which the rate of spin relaxation by SO coupling depends on the nanofilm thickness. To date, most measurements concerning SO coupling are performed far beyond the superconducting state, making a direct demonstration of how SO coupling affects superconductivity unavailable. In our experiments, when $T > T_c$, a WAL peak in the magneto-resistivity due to the presence of SO coupling is observed in all the nanofilms. By probing the temperature evolution of this resistivity peak toward the superconducting state, we have demonstrated that WAL effects are progressively suppressed as a result of the onset of superconductivity. In the superconducting state, the peak shape is clearly affected by Cooper pairing. Moreover, an unusual positive contribution to the magneto-resistivity (unexpected within the BCS theory and WAL effects) appears near the resistivity peak for the 3-nm-thick Al film in which the relaxation of spin is fastest among the three studied films. Our experimental work therefore highlights the important effects of SO coupling on superconductivity.
SO coupling in the normal state. Figure 1(a) shows the optical image of the studied 6-nm-thick sample and Fig. 1(b) shows the obtained longitudinal resistivity $\rho_{xx}$ over a wide range of temperature (5 K $\leq T \leq 30$ K). For $T > 18$ K, $\rho_{xx}$ increases with increasing $T$, consistent with the characteristics of a metal film subject to electron-phonon scattering. For $T < 18$ K, quantum corrections primarily due to WAL and superconducting fluctuations would then instead govern the $T$ dependence of resistivity as the effects of electron-phonon scattering is suppressed. Figure 1(c) shows the magneto-resistivity measurements for $0 < B < 11.7$ T at $T = 4.4$ K. We can observe that $\rho_{xx}$ is much larger than the Hall resistivity $\rho_{xy}$ over the whole field range. The smallness of $\rho_{xy}$ is due to a large carrier density in metal systems. Therefore one is able to obtain the longitudinal conductivity of $\sigma_{xx} \approx 1/\rho_{xx}$ according to $\sigma_{xx} = \rho_{xx}(\rho_{xx}^2 + \rho_{xy}^2)^{-1/2}$.

Figures 1(d)–(f) show $\rho_{xx}(B)$ at various $T$. As presented in Fig. 1(e), for 8 K $\leq T \leq 14$ K, signatures of WAL effects, that is, positive magneto-resistivity (MR) around $B = 0$ followed by negative MR with increasing $B$, are observed, clearly indicating the importance of SO coupling in the measured Al nanofilm with thickness of $d = 6$ nm. For $T \approx 16$ K, weak localization (WL) instead of WAL dominates the transport process, inferred from the monotonically decreasing trend of $\rho_{xx}$ with the applied $B$ shown in Fig. 1(d). Suppression of WAL at elevated $T$ is expected to occur when no significant spin-dependent phases can be accumulated. In metals, the dominant mechanism of SO coupling is believed to arise from the potential of lattice ions such that the spin orientation is randomized following the Elliot-Yafet (EY) mechanism which states that spin flips during every lattice ion scattering event.$^{19,25}$ According to the WAL theory developed by Hikami, Larkin, and Nagaoka (HLN) (ref. 26), which assumes that the EY mechanism is responsible for spin relaxation, the MR in the 2D diffusive regime under $\sigma_{xx} \approx 1/\rho_{xx}$ has the form

$$\frac{1}{\rho_{xx}} = \sigma_{xx}^0 + \frac{e^2}{2\pi^2 h} \left\{ \frac{1}{2} \left[ \Psi\left( \frac{1}{2} + \frac{B}{B_3} \right) - \Psi\left( \frac{1}{2} + \frac{B}{B_2} \right) - \frac{1}{2} \left[ \Psi\left( \frac{2}{B} \right) - \Psi\left( \frac{1}{B} \right) - \log \left( \frac{B_2^{1/2}}{B_1 B_3^{1/2}} \right) \right] \right\},$$

where $\sigma_{xx}^0$ is the zero-field conductivity independent of $B$, $\Psi(x)$ is the digamma function, $B_1 = B_0 + B_{SO}$, $B_2 = B_0 + \frac{4}{3} (B_{SO})$, and $B_3 = B_0$.

Here the characteristic fields $B_0$, $B_{SO}$ and $B_0$ are related to the elastic scattering length $l_0$, spin-orbit relaxation length $l_{SO}$, and dephasing length $l_\phi$ through $B_0 = \frac{h}{2el_0}$, $B_{SO} = \frac{h}{4l_{SO}}$, and $B_0 = \frac{h}{4l_\phi}$. The red curves in Figs. 1(d)–(f) denote the best fits to the data using Eq. (1). The excellent agreement between experiments and HLN theory shown in Figs. 1(d) and 1(e) for 8 K $\leq T \leq 24$ K suggests that the EY process is indeed the dominant mechanism for spin relaxation. However, it is found in Fig. 1(f) that HLN theory fails to describe the MR behavior for $T < 6$ K, which is ascribed to the influence of fluctuating Cooper pairing (superconducting fluctuations). The inset of Fig. 1(f) presents the fitting result considering SO interaction and Maki-Thompson superconducting fluctuations. A good agreement can likewise be observed. However such a method is not applicable as the superconducting state is well developed.

SO coupling in the superconducting state. In order to study the interplay between SO coupling and superconducting fluctuations around $B = 0$, we limit the fitting range to high magnetic fields, in which superconductivity is completely suppressed, to obtain the SO relaxation and dephasing length. Here we choose the field $B_0$ at which $\rho_{xx}(B)$ reaches its maximum as the lower bound to fit our data to Eq. (1). The underlying reason for this choice is that superconductivity cannot give rise to negative MR. On the other hand, the upper limit of this fit is chosen at $B = 2$ T to avoid the influence of positive MR which occurs at high fields due to orbital motion as shown in Fig. 1(c). The new fits to the data are shown as the red curves in Fig. 2(a) for 3 K $\leq T \leq 6$ K and further in Fig. 2(b) for 0.328 K $\leq T \leq 1.980$ K, where remarkable agreement between theory and experiments is found. Three important length scales $l_0$, $l_{SO}$, and $l_\phi$ can be determined by using the fit parameters and the results are plotted against $T$ in Figs. 3(a) and 3(b). We observe that $l_{SO}$ exceeds $l_\phi$ for $T \geq 14$ K, indicating that phase coherence has lost before spin-dependent phase is accumulated for $T \geq 14$ K. Consistently, WAL effects are barely observable for $T > 14$ K in Fig. 1(d). As shown in
the inset of Fig. 2(b), the zero-resistivity superconducting state appears when $T$ is low enough. The characterization of the 6-nm-thick superconducting nanofilm which yields important physical quantities can be found in the supplementary information. To further study the effects of Cooper pairing, we plot the theoretical curves given by Eq. (1) around $B = 0$ with the parameters determined from the fitting procedure at high $B$ as described above. These simulation curves, which are the extrapolation of the fitting curves, are represented by the blue ones in Fig. 2(a). The expected influence of superconductivity on WAL can then be tracked as a function of $T$ even down to 0.328 K, well inside the superconducting regime. By plotting $\rho_{xx}(T)$ at different $B$ in Fig. 2(c), we observe that there is a crossover between superconducting and insulating states with $B$.

By comparing the experimental data (where both SO and superconductivity exist) with the simulated curves (where only SO is considered) as shown in Figs 2(a) and 2(b), we observe that the position of resistivity peak, characterized by strong SO coupling in the normal state, is shifted to a higher magnetic field compared with the expected position when $T \approx 6$ K. Figure 3(c) then collects the difference between the measured and theoretical peak positions in their resistivity and magnetic field values at various $T$ covering both the normal and superconducting states. This difference becomes significant at low $T$, suggestive of suppression of WAL effects by spin-singlet Cooper pairing.

As shown in Fig. 2 (b), in the normal state indicated by the red fitted curves, $\rho_{xx}$ decreases with increasing temperature, characteristic of a weak insulator. When $B$ is below $B_p$, positive magneto-resistivity is observed and insulating behavior persists before the resistivity of the device reduces dramatically due to superconductivity. Since the $\rho_{xx}$ maxima predicted by the standard HLN model (indicated by the blue curves) occur at a substantially lower $B$ than those observed in experiments, $\rho_{xx}$ for $B < B_p$ cannot be solely described by spin-orbit coupling in the normal state and thus must be related to superconductivity. Such results are reminiscent of the intriguing influence of disorder on superconductivity. In a disordered superconductor subject to a strong perpendicular magnetic field, insulating behavior can occur. For a moderate amount of disorder in the system, the observed insulating behavior at high fields is
generally ascribed to localization of unpaired electrons. However, in a strongly disordered superconductor, increasing magnetic field does not always quench superconductivity\(^{34-36}\). Instead, thanks to the formation of superconducting islands (SCIs), with the presence of strong disorder, an applied out-of-plane magnetic field, which suppresses the correlations between different superconducting islands, is not detrimental to superconductivity drastically. The route toward insulating behavior with increasing magnetic field thereby proceeds through development of well-separated superconducting islands followed by localization of unpaired electrons, which is unexpected within the conventional BCS theory. Such an unusual superconductor-insulator transition (SIT) has already been observed in a wide variety of amorphous superconducting thin films including In\(_x\)O\(_y\) (ref. 27), TiN (ref. 29), and Bi (ref. 31). Strong disorder, which is believed to be detrimental to superconductivity, plays a crucial role in maintaining superconductivity in such systems. In the presence of strong disorder, normal-state transport should be governed by activation or hopping process, giving rise to an exponential dependence of resistivity value expected for the onset of strong localization.

For clarity, we plot \(\rho_{xx}(B)\) at \(T = 8\) K, 4.2 K, 3 K, and 2 K separately in Figs. 4(c) – (f), where Cooper pairing appears to cause the measured resistivity to be lower than that predicted by Eq. (1) due to stronger Cooper-pair correlations. Such results indicate that there will be a mechanism relevant to Cooper pairing which makes the resistivity higher than that predicted by Eq. (1).

The enlargement of SCI size with decreasing \(B\) is thought to be one of the mechanism which can provide an extra resistivity contribution. With a decrease in \(B\), the enlarged SCIs may further block the electron transport. As observed in Fig. 4(b), even for the 3-nm-thick film, the \(T\)-dependence of \(\rho_{xx}\) at \(B = 2\) T shown in Fig. 2(b). Moreover, the obtained \(\rho_{xx}\) over the whole measurement range is far below \(\frac{\hbar}{2e^2} \approx 13\) k\(\Omega\) which is the critical resistivity value expected for the onset of strong localization.

Further magneto-transport measurements were performed on the Al nanofilms with thickness of 3 nm and 12 nm, which are expected to have respectively faster and slower spin relaxation compared with that of 6 nm. Figures 4(a) and 4(b) show the measured \(\rho_{xx}(B)\) at various \(T\) for the 3-nm-thick film. We observe that positive MR around \(B = 0\) due to WAL, can be well fitted to Eq. (1) for \(T \approx 8\) K. For \(T < 8\) K, influence of superconductivity on the magneto-resistivity becomes increasingly important with decreasing \(T\). For clarity, we plot \(\rho_{xx}(B)\) at \(T = 5\) K, 4.2 K, 3 K, and 2 K separately in Figs. 4(c) – (f). The experimental data are in good agreement with WAL theory described by Eq. (1) when \(B > B_p\), the field where the resistivity peak occurs. Following the same analysis procedure done for the 6-nm-thick film, we extrapolate the high-field fitting curves to lower fields by directly substituting the extracted fitting parameters into Eq. (1). Interestingly, as seen by the comparison between measured and simulated curves for \(B < B_p\) superconductivity does not always cause the measured resistivity to be lower than that predicted by Eq. (1) for a given \(B\). For example, figure 5(c) shows that at \(T = 5\) K the magneto-resistivity near \(B_p\), is higher than its expected value without taking superconducting effects into account given by Eq. (1). With decreasing \(T\), this unusual positive resistivity contribution becomes gradually smaller and eventually disappears, which is observed clearly in the insets of Figs. 4(c) – (f), the zoom-in of the region around \(B_p\). At lower \(T\) and \(B\), the resistivity value is always lower than the prediction of Eq. (1) due to stronger Cooper-pair correlations. Such results indicate that there will be a mechanism relevant to Cooper pairing which makes the resistivity higher than that predicted by Eq. (1).

**Discussion**

The physical quantities of superconductivity and SO coupling obtained from the fits for the Al nanofilms of various thicknesses are summarized in Table 1. We note that \(\zeta\) is longer than the film thickness in all our samples. Therefore we always have quasi-2D superconductivity. We can clearly see in Table 1 that \(t_{SO} (= k_{SO}^2/D)\) (ref. 38) decreases with decreasing the film thickness, indicating
that the strength of spin relaxation is stronger in a thinner film. Correspondingly, the amount of disorder increases with decreasing the film thickness, which can be understood from the comparison of the zero-field resistivity at $T = 8$ K well above $T_c$ and from the temperature dependence of resistivity $\rho(T, B = 0)$ for $T \approx 8$ K (see the Supplementary Fig. S1). We can see clearly that the insulating behavior becomes progressively stronger with reducing the film thickness. It is known that two extensively studied effects of spin relaxation are the Elliot-Yafet and Dyakonov-Perel (DP) mechanisms\textsuperscript{25,29,40}. In the EY mechanism, spin relaxation is relevant to how the spin changes its orientation during a scattering process. This is possible as the electron wave functions associated with a given spin acquire an admixture of the opposite-spin state due to SO coupling induced by lattice ions. In contrast, the DP mechanism is related to the Dresselhaus SO coupling caused by bulk inversion asymmetry and the Rashba SO coupling caused by structural inversion asymmetry. In this case, a charged particle moving in an electric field (originating from the lack of inversion symmetry) experiences an effective magnetic field, which couples to its spin. The spin relaxation is thereby relevant to spin precession around this effective magnetic field between scattering events. Therefore, increasing disorder may enhance the rate of spin relaxation governed by EY mechanism. Moreover, as shown in the Supplementary Fig. S2, the model developed by Iordanskii, Lyanda-Geller and Pikus (ILP) (ref. 41), in which the DP mechanism is included, cannot yield good fits to the measured MR data as compared to the HLN model especially in the 3-nm-thick film with the strongest disorder in our experiments. These results indicate that EY mechanism dominates over DP one with decreasing the nanofilm thickness.

The scale of energy variation involved due to spin relaxation can be evaluated utilizing the uncertainty relation $e \Delta = h/\tau_{\text{SO}}$ (ref. 14). By calculating the ratio of $e \Delta$ to $2A$, which is found to be 0.37, 0.77, and 0.85 for the films of $d = 12$ nm, 6 nm, and 3 nm, respectively, the growing importance in the correlation between spin relaxation and superconductivity with decreasing the film thickness is thereby revealed. Correspondingly, $l_{\text{SO}}$ approaches progressively to $\xi$. This numerical analysis suggests that the unexpected resistivity contribution around $B_p$ observed in the 3-nm-thick film is indeed related to the combination effects of SO coupling which causes spin relaxation and superconductivity which pairs electron spins.

Figure 5 | (a) & (b) Magneto-resistivity measurements $\rho_{xx}(B)$ on the 12-nm-thick film at different temperatures $T$. The red curves correspond to the best fits to Eq. (1).

Table 1 | Summary of the physical quantities of superconductivity and spin-orbit coupling

| $d$ (nm) | $T_c$ (K) | $2A$ (meV) | $H_c(0)$ (T) | $\xi$ (nm) | $D$ (10^{-4} m^2/s) | $\ell^{\text{so}}$ (nm) | $\tau_{\text{SO}}$ (10^{-11} sec) | $\varepsilon_{\text{SO}}$ (meV) | $\rho_{\text{me}}(T = 8$ K, $B = 0)$ | $\varepsilon_{\text{me}}/2\Delta$ | $l_{\text{me}}/\xi$ |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 3 | 1.509 | 0.46 | 0.339 | 31.18 | 2.44 | 51.30 | 1.08 | 0.39 | 1099 | 0.85 | 1.65 |
| 6 | 1.577 | 0.48 | 0.424 | 27.88 | 2.04 | 48.12 | 1.13 | 0.37 | 154 | 0.77 | 1.73 |
| 12 | 1.358 | 0.41 | 0.139 | 48.69 | 5.36 | 120.90 | 2.72 | 0.15 | 22 | 0.37 | 2.48 |

* $\ell^{\text{so}}$ shown here is obtained by averaging the extracted parameters below 4.5 K which are normally T-independent.
close to the MR peak occurring accompanied by the appearance of superconductivity in the 3-nm-thick film having faster spin relaxation. For the 12-nm-thick film, since superconducting correlation is rather strong and spin relaxation is comparably slower, MR peak is completely suppressed at low $T$. Different behavior in the films of different thicknesses results from different strength of spin relaxation and superconductivity. Spin-orbit coupling, which makes the spin degree of freedom respond to its orbital characteristics and causes spin relaxation, has thereby been demonstrated to have important effects on superconductivity. Our experimental work sheds new light on the understanding of the role of SO coupling in a superconductor.

Methods

All samples were grown in situ using a Varian Gen II solid-source molecular beam epitaxy (MBE) system. For an Al film whose thickness is $30–100$ nm, an undoped 200-nm-thick GaAs buffer layers is grown on semi-insulating (100) GaAs substrate as a template. We find that the thickness of the GaAs buffer layer needs to be increased to 300 nm to achieve a flat enough template for depositing an aluminum nanofilm which is thinner than 15 nm. Before Al deposition, the buffer layer on top of the semi-insulating (100) GaAs substrate is kept in the ultra-high vacuum to prevent the GaAs surface from oxidation. After the growth chamber is pumped down to $3 \times 10^{-10}$ Torr to eliminate residual arsenic vapor pressure, an Al nanofilm is grown at ~0°C at a slow rate of 0.18 $\mu$m/h ($0.05$ nm/s) (ref. 42). We note that once the film thickness is below 2 nm, the Al nanofilm does not conduct at room temperature, suggesting that an ultrathin Al film gets oxidized and become Al$_2$O$_3$, which appears to be an insulator. For an Al nanofilm whose as-grown thickness is $\geq 3$ nm, the top Al$_2$O$_3$ film serves as a protective layer so that the underneath Al nanofilm does not get oxidized during processing. All samples were processed into 50-μm-wide Hall bars using conventional photo-lithography and lift-off technique. The mesa is defined by 2% tetra-methyl ammonium hydroxide (TMAH) etching for 20 seconds for phasing in two-dimensional electron system transferring from photore sist to aluminum film. It is found that such a slow etch rate is crucial for high-yield, manageable and reliable sample preparation. A 30 nm/300 nm Ti/Au layer is deposited using electron-gun evaporation as the contact electrodes. Four-terminal magneto-transport measurements were carried out using dc constant-current sources in a top-loading He$^+$ cryostat equipped with a superconducting magnet. Since the device was patterned into Hall bar geometry one can obtain both longitudinal and Hall resistances $R_{xx}$ and $R_{xy}$. In Fig. 3(a), we note that the mean free path $l_0$ is larger than the thickness (~6 nm) of the film being measured, indicating that the nanofilm in the present study can be treated as a quasi-two-dimensional system. Here we can define longitudinal and Hall resistivities $\rho_{xx}$ and $\rho_{xy}$ by $R_{xx}=\rho_{xx}wl$ and $R_{xy}=\rho_{xy}$, where $w$ is the width of the Hall bar and $l$ is the distance between the two voltage probes used for the measurement of $R_{xy}$. The same results were obtained on 3-nm and 12-nm-thick films. Source-drain current $I_{SD}$ on the order of 10 μA was injected to improve the signal-to-noise ratio. For $I_{SD} < 1$ μA, the weak localization effects were hardly observed, although large current $I_{SD}$ cause heating effects, which increases the electron temperature, our main findings regarding the coupling of spin-orbit coupling and superconductivity would not be influenced since $I_{SD}$ is not large enough to completely destroy the superconducting state.

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Author contributions
S.T.L., S.W.L. and Y.T.W. performed the measurements. S.W.L. and S.D.L. prepared the samples. S.T.L. and C.T.L. drafted the paper. S.D.L. and C.T.L. coordinated the project.

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