Elliott-Yafet Spin-Orbit Scattering in Ultrathin Semimetallic SrIrO$_3$ Films

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Abstract

We report a magnetotransport study of spin relaxation in high quality epitaxial SrIrO$_3$ thin films coherently strained on SrTiO$_3$ substrates. Fully charge compensated semimetallic transport has been observed in 2.0-21.2 nm SrIrO$_3$ films, and the thinner films exhibit insulating behavior. For metallic samples, the charge mobility at 10 K increases from 45 cm$^2$/Vs to 141 cm$^2$/Vs with decreasing film thickness. In the two-dimensional regime, the charge dephasing and spin-orbit scattering lengths extracted from the weak localization/anti-localization effects show power-law dependence on temperature, pointing to the important role of electron-electron interaction. While the spin-orbit scattering time $\tau_{so}$ in the thicker films exhibits an Elliott-Yafet mechanism dominated quasi-linear dependence on the momentum relaxation time, ultrathin films approaching the critical thickness of metal-insulator transition show about 6 times higher $\tau_{so}$, which has been attributed to the onset of enhanced electron correlation in SrIrO$_3$ close to the charge gap opening.
The Ruddlesden-Popper series iridates \((\text{Sr}_{n+1}\text{Ir}_n\text{O}_{3n+1})\) have drawn considerable research interests in recent years due to the strong spin-orbit coupling (SOC), which competes with the electron itineracy, the on-site Coulomb energy \(U\), and the crystal fields associated with the spatially extended \(5d\)-orbitals.\(^{1-4}\) As the end member of the series in the large bandwidth limit \((n = \infty)\), \(\text{SrIrO}_3\) (SIO) is a paramagnetic semimetal,\(^{3-5}\) and has been suggested to exhibit nontrivial topological phases, such as Weyl/Dirac semimetals and topological Mott insulators, in the orthorhombic phase with space group \(Pbnm\) [Fig. 1(a)].\(^6,^7\) While the perovskite SIO is metastable in bulk,\(^8,^9\) it has been realized in high quality epitaxial thin film form,\(^4,^{10}\) where a range of interesting phenomena have been observed, including strain tunable Dirac node\(^{11-13}\) and metal-insulator transition (MIT),\(^{14-16}\) and thickness-driven dimensionality crossover.\(^{17,18}\) Due to the superb conduction and intrinsically large SOC, SIO is a promising material candidate for developing Datta-Das spin field effect transistors.\(^{19}\) While the ability to control spin dephasing is critical for the practical implementation of this device concept, the dominant spin relaxation mechanism in ultrathin SIO films, especially in the presence of enhanced electron correlation, remains elusive to date.

In this work, we report a comprehensive magnetotransport study of spin relaxation in high quality epitaxial SIO thin films coherently strained on \(\text{SrTiO}_3\) (STO) substrates. SIO films with thickness down to 2.0 nm show semimetallic transport with complete charge compensation, with thinner films exhibiting insulating behavior. From the two-dimensional (2D) weak localization (WL) and weak anti-localization (WAL) effects, we extracted the phase coherence length and spin-orbit scattering length as functions of temperature and film thickness. In thicker films, the spin-orbit scattering time \(\tau_{so}\) exhibits a quasi-linear dependence on the momentum scattering time, pointing to Elliott-Yafet (EY) mechanism dominated spin relaxation. The films
approaching the critical thickness of MIT show about 6 times higher \( \tau_{so} \), with concomitantly reduced carrier density and increased charge mobility. The observed effects can be attributed to the enhanced correlation in ultrathin SIO close to the charge gap formation, which suppresses spin scattering.

We deposited 1.4-21.2 nm epitaxial SIO thin films on (001) STO (1.4% compressive strain) using off-axis radio frequency magnetron sputtering (Supplemental Material).\(^{20}\) Atomic force microscopy (AFM) measurements reveal smooth sample morphology with typical surface roughness of 1-2 Å [Fig. 1(b)]. X-ray diffraction (XRD) spectra show (001) growth (pseudo-cubic notation) with no appreciable impurity phases [Fig. 1(c)]. The \( c \)-axis lattice constant is \( \sim 4.02 \) Å, which is larger than the bulk value of 3.96 Å and consistent with that reported for strained SIO on STO.\(^{15}\) Reciprocal space mapping (RSM) shows that even the thickest film is fully strained to the substrate [Fig. 1(d)]. The films were then fabricated into Hall bar devices for magnetotransport studies, with details given in the Supplemental Material.\(^{20}\)

Figure 1(e) shows the temperature dependence of sheet resistance \( R_\square(T) \) taken on the SIO films. For the 2.0 nm and thicker films, we observe metallic temperature dependence \((dR/dT > 0)\) over a wide temperature range, followed by a natural logarithmic \( T \)-dependence of sheet conductance \( G \) below a critical temperature \( T_m \):\(^{20}\)

\[
R_\square = R_0 + A \cdot T^\alpha \quad T > T_m \quad (1a), \text{ and}
\]

\[
G = G_0 + \frac{e^2}{\pi h} \ln(T/T_0) \quad T < T_m \quad (1b).
\]

Here \( R_0 (G_0) \) is the residual sheet resistance (conductance), and \( \alpha, p \) and \( T_0 \) are fitting parameters. Distinct from the hexagonal SIO single crystals,\(^5\) the high temperature \( R_\alpha(T) \) exhibits very weak temperature dependence, with room temperature resistivity \( \rho(300 \, K) \) of \( 0.5 - 0.9 \) mΩ cm. The power exponent \( \alpha \) in Eq. (1a) is about 1.1-1.2,\(^{20}\) which reveals a non-Fermi-liquid behavior. The
low temperature $\ln(T)$ correction to the conductance is the signature behavior of a 2D electron system [Eq. (1b)], which can originate from either WL or electron-electron interaction.\textsuperscript{21} As the film thickness is further reduced, SIO becomes insulating over the entire temperature range [Fig. 1(e)]. The low temperature $R_{\square}(T)$ for the 1.6 nm film can be well described by the 2D variable range hopping (VRH) model: $R_{\square} \propto \exp \left( \frac{T_0}{T} \right)^{1/3}$.\textsuperscript{20, 22} The transition to the strongly localized behavior occurs as $R_{\square}$ exceeds $h/2e^2 \approx 12.9 \, \text{k}\Omega$. The 1.4 nm film, on the other hand, exhibits a crossover from thermally activated semiconducting behavior $\exp[E_a/2k_BT]$ at high temperature to 2D VRH at low temperature,\textsuperscript{20} with the extracted activated energy $E_a \sim 100 \, \text{meV}$ consistent with previous reports for ultrathin SIO.\textsuperscript{17} Such thickness-driven MIT in SIO has been attributed to enhanced correlation energy in the 2D limit\textsuperscript{17} and lattice distortion imposed by the substrate symmetry.\textsuperscript{18} The film thickness where the insulating behavior emerges (about 4 unit cell) is comparable with that reported for SIO films encapsulated with STO top-layers,\textsuperscript{17} and approaches the critical thickness for the charge gap formation,\textsuperscript{18} further attesting to the high quality of our samples.

The semimetallic nature of SIO is clearly manifested in the Hall effect and magnetoresistance (MR) measurements. Figures 2(a)-(b) show the Hall resistance $R_{xy}$ and $R_{\square}$ of the 2.0 nm SIO as functions of magnetic field $H$, respectively, which can be well described by the semiclassical two-band model,\textsuperscript{23}

\begin{equation}
\rho_{xy} = \frac{(n \mu_e^2 - n \mu_h^2)H + \mu_e^2 \mu_h^2(p-n)H^3}{e(n \mu_e + p \mu_h)^2 + (p-n)^2(\mu_e \mu_h)^2} \tag{2a}\end{equation}

\begin{equation}
\frac{\Delta \rho}{\rho(0)} = \frac{\rho(H)-\rho(0)}{\rho(0)} = \frac{(n \mu_e + p \mu_h)^2 + \mu_e \mu_h(n \mu_e + p \mu_h)(p \mu_e + n \mu_h)H^2}{(n \mu_e + p \mu_h)^2 + (p-n)^2(\mu_e \mu_h)^2} - 1 \tag{2b}.
\end{equation}

Here $\rho_{xy} = R_{xy} t$ is the Hall resistivity, $t$ is the film thickness, $\rho$ is the longitudinal resistivity, $n$ ($p$) is the electron (hole) density, and $\mu_e$ ($\mu_h$) is the electron (hole) mobility. The linear $H$-
dependence of $R_{xy}$ has been observed in films of all thicknesses, which implies that the electron and hole densities are fully compensated, i.e., $n \approx p$ [Fig. 2(a)]. Fitting the Hall and MR data to Eq. 2 and assuming $n = p$, we extracted the carrier density and mobility as functions of film thickness. At $T_m$, the carrier density falls in the range of $10^{19}$ and $10^{20}$ cm$^{-3}$ [Fig. 2(c)], similar to previously reported results.$^{15,16}$ At 10 K, the 2.0 nm and 2.8 nm films show considerably lower carrier density than those at $T_m$, consistent with an emerging charge gap at the ultrathin limit.$^{17,18}$ The carrier mobility, on the other hand, is significantly enhanced in the thinner films [Fig. 2(d)], with $\mu_e$ at 10 K increasing from 45 cm$^2$/Vs in the 21.2 nm film to 141 cm$^2$/Vs in the 2.0 nm film. The mobility difference for electron and hole, defined as $\beta = \frac{\mu_e - \mu_h}{\mu_e + \mu_h}$, is less than 1.5% for all samples. The electron mobility ($\mu_e$) is slightly higher than the hole mobility ($\mu_h$), which may be due to the heavier hole mass.$^{11,13}$ The mobility values are orders of magnitude higher than those observed in strongly correlated oxides in the paramagnetic metallic phase,$^{24}$ and comparable with the non-interacting electrons in thin films of high mobility perovskite semiconductor BaSnO$_3$.$^{25}$ A possible scenario is these samples are close to the fully charge compensated regime with low density of states in both the conduction and valence bands,$^{11}$ which suppresses the intra-band elastic scattering. The inter-band scattering does not play a critical role at low temperature due to the large momentum transfer required between the electron and hole bands at the Fermi level $E_F$.$^{11,13}$

Figure 3(a) shows the magnetoconductance (MC) $\Delta \sigma(H) = \sigma(H) - \sigma(0)$ of the 2.8 nm SIO film at low temperatures. The sample exhibits a negative MC at low field followed by a positive MC at high field. The positive MC is widely observed in 2D electron systems due to the WL effect, where the magnetic field suppresses the constructive back scattering of electrons. The negative MC, known as WAL, originates from spin-orbit scattering, which leads to destructive
interference of back-scattered electrons that can be suppressed by the magnetic field. The WAL
effect has previously been used to deduce SOC induced spin splitting in semiconductor
heterostructures.\textsuperscript{26, 27} In SIO films, the transition from WL to WAL dominated MC occurs at
progressively higher field at increasing temperatures, suggesting that the dephasing/scattering
mechanisms contributing to these two quantum conductance correction effects have different
temperature-dependences.

Given the strong SOC in SIO and considering the effect of Zeeman splitting, we employed
the Maekawa-Fukuyama (MF) model to fit $\Delta \sigma(B)$:\textsuperscript{28}

$$
\frac{\Delta \sigma(H)}{\sigma_0} = \Psi \left( \frac{H}{H_i + H_{so}} \right) + \frac{1}{2\sqrt{1-\gamma^2}} \Psi \left( \frac{H}{H_i + H_{so} \left(1 + \sqrt{1-\gamma^2}\right)} \right) - \frac{1}{2\sqrt{1-\gamma^2}} \Psi \left( \frac{H}{H_i + H_{so} \left(1 - \sqrt{1-\gamma^2}\right)} \right)
$$

(3).

Here $\Psi = \ln(x) + \psi(1/2 + 1/x)$, $\psi(x)$ is the digamma function, and $\sigma_0 = e^2/\pi h$ is the
quantum conductance. The dephasing driven by inelastic scattering and spin-orbit scattering are
captured in $H_i = \frac{h}{4eD\tau_i}$ and $H_{so} = \frac{h}{4eD\tau_{so}}$, respectively, where $D = \frac{1}{2}L_p\tau_p$ is the diffusion constant,
$L_p$ is the elastic mean free path, $\tau_p$ is the momentum relaxation time, and $\tau_i$ is the inelastic
scattering time. The parameter $\gamma = g\mu_B H/4eDH_{so}$ couples $H_{so}$ and the $g$ factor, which describes
the Zeeman effect correction. At this temperature range (2-5 K), the MC data of the 2.8 nm SIO
can be well described by the MF model [Fig. 3(a)]. Given the sample mobility ($\mu_{e,h} \sim 140$ cm\textsuperscript{2}/Vs
at 10 K), the criterion of low magnetic field condition ($\mu_{e,h}B < 1$) is satisfied in the entire range
of magnetic field investigated. The quantum corrections to the MC persists to very high field (up
to 4 T), while the classical parabolic contribution is negligible.

Figure 3(a) inset shows the temperature dependence of the extracted $H_i$ and $H_{so}$. While $H_i$
increases linearly with increasing temperature, $H_{so}$ exhibits a much stronger, quadratic $T$-
dependence. Similar temperature dependence has been observed in the 2.0-5.4 nm samples,
while thicker films do not show prominent WL and WAL in MC above 3 K. From $H_i$ and $H_{so}$ we extracted the inelastic scattering and spin-orbit scattering lengths using $L_{i,so} = (D\tau_{i,so})^{1/2} = \left(\frac{\hbar}{4eH_{i,so}}\right)^{1/2}$. As shown in Fig. 3(b-c), both scattering lengths exhibit power-law dependences on temperature $L_{i,so} \sim T^\alpha$. For $L_i$, $\alpha = 0.4 \pm 0.1$, close to the expected value of $\alpha = 0.5$ for electron-electron scattering induced dephasing.21 The exponent for $L_{so}$ is $\alpha = 1.0 \pm 0.1$, in sharp contrast to the nearly $T$-independent $L_{so}$ observed in semiconductor quantum wells,27 where spin-orbit scattering is related to the spin precession process induced by the intrinsic spin-splittings in the 2D band.

Figure 4(a) summarizes the thickness dependence of $L_i$ and $L_{so}$ for the 2.0-10.2 nm SIO films at 2 K. While $L_i$ does not exhibit any apparent thickness dependence, $L_{so}$ decreases progressively in thicker films. We can thus rule out the film surface/interface states as the major electron dephasing/spin scattering source. The $L_i$ value is always higher than $L_{so}$, which satisfies the strong spin-orbital scattering condition in the MF model, i.e., $\frac{\tau_i}{\tau_{so}} > 0.183$,28 justifying the observation of WAL at low field and WL at high field.

To confirm that these samples are in the 2D diffusive region, we calculated the elastic mean free path in these samples. Previous band structure mapping via angle-resolved photoelectron spectroscopy has revealed a light mass electron pocket and a heavy mass hole pocket at $E_F$.11,13 Given the significantly enhanced hole mass (~6 $m_0$ with $m_0$ the free electron mass), the quantum correction to conduction can only be manifested in the electron transport at the temperature and field range investigated. We thus estimated the electron mean free path using the Drude model

$L_p = v_F\tau_p$, with $\tau_p = \mu_e m^*/e$, $m^*$ the effective electron mass, $v_F = \frac{\hbar k_F}{m^*}$ the Fermi velocity, $k_F = \sqrt{2\pi n_{2D}}$ the Fermi wave vector, and $n_{2D} = n_e t$ the 2D electron density. Considering that
the Dirac node is lifted by the compressive strain, we assumed a parabolic band with $m^* \approx m_0$.\textsuperscript{11-13} This assumption is supported by the observed carrier density, which is significantly higher than what is expected for a linear dispersion.\textsuperscript{9, 11, 20} In Fig. 4(a), $L_p$ increases moderately with film thickness, which can result from the increasing carrier density in thicker films [Fig. 2(c)]. We used the condition of $L_p$ exceeding the film thickness to gauge if the system can be characterized as 2D. While the 10.2 nm sample sits right at the $L_p = t$ boundary, the thinner films reside well within the 2D regime, confirming the thickness-driven dimensionality crossover. Nevertheless, the condition for the WAL regime, $L_p^2 < L_{so}^2 \leq L_i^2$, is always satisfied for the electrons.\textsuperscript{28} The magnetic length at 4 T $L_B = \sqrt{\hbar/eB} \approx 13$ nm also exceeds $L_p$, confirming that the system is in the diffusive regime for the entire field range.

We consider two commonly observed spin relaxation mechanisms: the D’yakonov-Perel’ (DP) type and the Elliot-Yafet type.\textsuperscript{29} The DP mechanism describes the spin precession induced by the spin splitting of the energy level that’s randomized by the elastic scattering,\textsuperscript{29-31} and can have both the bulk (Dresselhaus) and interface (Rashba) contributions. The bulk term is important in noncentrosymmetric materials.\textsuperscript{26} The Rashba effect is due to the inversion field at the hetero-interfaces and surfaces, as observed in semiconductor quantum wells\textsuperscript{27} and complex oxide heterostructures,\textsuperscript{32, 33} and is expected to be more prominent in thinner films. The EY mechanism, on the other hand, depicts the spin dephasing via momentum scattering, e.g., due to impurities or electron-phonon interactions, in the presence of SOC.\textsuperscript{29, 34, 35} Structural defects commonly occurred in epitaxial thin films, such as cation vacancies, dislocations, and grain boundaries, can all contribute to the spin scattering through the EY mechanism. One way to distinguish these two mechanisms is to examine the relation between $\tau_{so}$ and $\tau_p$.\textsuperscript{29} In the DP mechanism, the SOC induced spin splitting acts effectively as a magnetic field that causes
electron precession. The elastic scattering randomizes this process, so that the related spin dephasing time scales with $1/\tau_p$.\textsuperscript{26} In the EY mechanism, in contrast, the spin-flip process is facilitated by momentum scattering. The associated $\tau_{so}$ is thus expected to depend linearly on $\tau_p$.\textsuperscript{36}

In SIO thin films, while the WL-WAL type measurements have previously been performed to extract $\tau_{so}$,\textsuperscript{14,16} the relation between $\tau_{so}$ and $\tau_p$ remains elusive due to the lack of effective approaches to control the charge mobility.\textsuperscript{11,14-16} For example, in the study of WAL in SIO thin films grown on LaAlO$_3$ substrates, only 1% variation of $\tau_p$ has been reported.\textsuperscript{14} The film thickness of our samples, on the other hand, presents a powerful control parameter to tune carrier mobility, making it feasible to identify the dominant spin relaxation mechanism in epitaxial SIO thin films. Figure 4(b) shows $\tau_{so}$ at 2 K as a function of $\tau_p$. To avoid the complicating effect of quantum conductance correction, we used the $\tau_p$ values deduced from mobility at 10 K. For the thicker films, $\tau_{so}$ exhibits a quasi-linear relation with $\tau_p$, increasing from 0.093±0.008 ps in the 10.2 nm film to 0.27±0.02 ps in the 4.2 nm film, which points to an EY mechanism dominated spin relaxation. The absence of Dresselhaus contribution is not surprising given that the orthorhombic structure of SIO preserves inversion symmetry. The carrier density and film thickness dependences of $H_{so}$ also rule out a dominant presence of the Rashba effect.\textsuperscript{20,26}

Next we discuss the possible spin scattering sources in epitaxial SIO films. As $\tau_{so}$ is enhanced in the thinner films, it is natural to rule out the roughness and defect states related to the film interface/surface as the dominant spin scattering sources. A possible bulk mechanism that contributes to the lower $\tau_{so}$ in thicker films is the structural defects associated with the epitaxial strain, whose density can increase progressively with film thickness. As orthorhombic SIO is meta-stable in bulk, it has been shown that the film crystallinity degrades in the thicker
films, where the lattice strain is released through oxygen vacancies, dislocations, and even formation of polycrystalline grains above a critical thickness (20-40 nm).\textsuperscript{4,10} The last mechanism is not relevant as our samples are well below the critical thickness, and XRD studies yield no sign of impurity phase growth. We also note that the charge mobility increases in thinner films, which possess lower carrier density [Figs. 2(c-d)]. This rules out a pronounced presence of charged impurities, e.g., cation or oxygen vacancies, as they are highly susceptible to the screening effect. On the other hand, misfit dislocations can present even in ultrathin films,\textsuperscript{14} and is a viable source for spin scattering.

The EY-mechanism also naturally accounts for the unusually strong temperature dependence of $\tau_{so}$ [Fig. 4(b) inset]. In the presence of a nearby band, spin flipping involves the electron-electron inter-band scattering,\textsuperscript{34,37} which can yield a $T^2$-dependent scattering rate.\textsuperscript{38} In fact, the $T^2$-dependence of resistivity has been observed in thick SIO films on GdScO$_3$ substrates below 10 K.\textsuperscript{15} In ultrathin films, this effect cannot be directly observed in low temperature $R(T)$ due to the conductance correction from WL and electron-electron interaction.\textsuperscript{21}

For the 2.0 nm and 2.8 nm films, $\tau_{so}$ exhibits an abrupt enhancement to 1.9±0.1 ps and 2.0±0.1 ps, respectively, which is 6 times higher than that of the 4.2 nm film. This change is accompanied by a sudden drop of the carrier density [Fig. 2(c)], consistent with the fact that these films are approaching the critical thickness for the charge gap formation.\textsuperscript{17,18} Similar enhancement in spin dephasing time close to the MIT has previously been observed in GaN.\textsuperscript{39} In the EY mechanism, the momentum scattering induces spin flipping between two neighboring bands with admixed spin-up and spin-down states. The emerging energy gap at $E_F$ can suppress the spin transition probabilities, leading to enhanced $\tau_{so}$.\textsuperscript{36} The abrupt change of $\tau_{so}$ vs. $\tau_p$ relation may also signal a crossover from EY to DP dominated regime\textsuperscript{40} due to the evolving
relative strengths of Rashba effect, uncertainty of the Bloch state energy $\frac{\hbar}{\tau_p}$, and correlation enhanced spin-orbit splitting in ultrathin films.\textsuperscript{41}

In summary, we report a weak (anti)localization study of spin relaxation in high quality epitaxial SrIrO$_3$ thin films. In the 2D regime, the spin-orbit scattering time in SIO exhibits quasi-linear dependence on the electron mobility in thicker films, pointing to the EY mechanism dominated spin relaxation, and is strongly enhanced in films close to the critical thickness of MIT. Our study reveals the complex interplay of SOC, impurity scattering, and electron correlation on spin transport in this emerging quantum material. The ultrathin films exhibit enhanced mobility and spin scattering time, making them an ideal platform for realizing novel spintronic devices such as spin transistors and SOC enhanced multiferroic tunnel junctions.

Acknowledgement:

We thank Allen MacDonald and Kirill Belashchenko for valuable discussions. This work was primarily supported by the National Science Foundation (NSF) through Grant No. DMR-1710461. Additional support was provided by the Nebraska Materials Research Science and Engineering Center (MRSEC) (NSF Grant No. DMR-1420645) (materials preparation). The research was performed in part in the Nebraska Nanoscale Facility: National Nanotechnology Coordinated Infrastructure and the Nebraska Center for Materials and Nanoscience, which are supported by the National Science Foundation under Award ECCS: 1542182, and the Nebraska Research Initiative.
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20 See Supplemental Material at xxx for detials of sample growth and characterization, device fabrication, and data modeling.

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Fig. 1. (a) Schematic of orthorhombic SIO. (b)-(d) Characterizations of a 21.2 nm SIO on STO. (b) AFM topography image shows smooth film surface with atomically flat terraces. (c) XRD θ-2θ scan with fits to the Laue oscillations around the Bragg peaks (dotted lines). (d) RSM around STO (103) peaks. (e) $R_\square(T)$ for films with different thicknesses. Inset: $R_\square(T)$ for the 2.0 nm film with fits to Eq. (1a) (dashed line) with $\alpha = 1.1$ and Eq. (1b) (dotted line) with $p = 0.85$. 
Fig. 2. (a) $R_{xy}$ and (b) $R_{\square}$ vs. $H$ taken on the 2.0 nm SIO at 20 K. The red lines are fits to Eq. 2. (c) Electron density and (d) mobility vs. film thickness at $T_m$ and 10 K.
Fig. 3. (a) MC of 2.8 nm SIO at various temperatures with fits to Eq. 3 (solid lines). Inset: $H_i$ and $H_{so}$ vs. $T$ with fits to $T$ (dashed line) and $T^2$ (dotted line) dependences, respectively. (b) $L_i$ and (c) $L_{so}$ vs. $T$ taken on films of various thicknesses. The dashed lines serve as the guide to the eye.
Fig. 4. (a) $L_i$, $L_{so}$ and $L_p$ vs. film thickness. The dotted line set at $L = t$ divides the 2D and 3D (shadowed area) regimes. The dashed lines serve as the guide to the eye. (b) $\tau_{so}$ at 2 K as a function of $\tau_p$. The dashed line is a linear fit. Inset: $\tau_{so}$ vs. $T$ for the 2.8 nm SIO with a fit to $T^2$ dependence (dotted line).