Large positive linear magnetoresistance in the two-dimensional $t_{2g}$ electron gas at the EuO/SrTiO$_3$ interface

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Supplementary Note 1

Supplementary Figure S6(a) displays XPS valence band spectra for the bare STO substrate (blue shading) and 10-nm-thick EuO film (red shading). Band offsets were calculated by both core-level\textsuperscript{1} and valence band\textsuperscript{2,3} spectroscopy. The valence band maximum ($E_{VBM}$) for each material was calculated using the linear extrapolation method.\textsuperscript{4} By comparing energy offsets between the Ti 2p, Eu 3d, and valence band edge positions (as summarized in Table 1) using

$$VBO_{CL} = \left( E_{Eu3d} - E_{VBM} \right)_{EuO} - \left( E_{Ti2p} - E_{VBM} \right)_{STO} - \left( E_{Eu3d} - E_{Ti2p} \right)_{EuO/STO}$$

we calculate a $VBO_{CL}$ of 2.44 eV.

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The VBO can also be determined directly from the valence band spectra.\(^2,^3\) For a heterostructure of 1 nm EuO/STO a contribution from the substrate is visible along with an attenuated contribution from the substrate (Supplementary Figure S6 (a), black open circles). We simulated a fit (purple solid line) to the heterostructure by scaling and offsetting the pure valence band spectra measured for the STO substrate and thick EuO film. This method uses an “all at once” fit minimizing \(\chi^2\) by the Levenberg-Marquardt algorithm implemented in Igor Pro software (WaveMetrics, Lake Oswego, OR). The difference between the measured and simulated spectra is also plotted (black line). Using this method, we calculate \(\text{VBO}_{\text{sim}} = 2.26\) eV, in good agreement with the offset calculated above by the core levels. The band profile is illustrated in Supplementary Figure S6(b).

**Supplementary Note 2**

Our calculations follow Onose et al.’s work\(^5\), which is based on Boltzmann transport theory. In the present case, we have one interface (IF) \(d^\uparrow_{xy}\) band, and three unpolarized \(t_{2g}\) bands for deep layers (we call them bulk \(t_{2g}\) states). All these states are confined along the \(z\) direction. From \textit{ab-initio} calculations, the IF band is found to be split down by \(\Delta\) compared with the other bands and \(\Delta\) is about 0.15 eV. Thus, we can write the energy near the \(I\) point for these bands\(^6\):

\[
\epsilon = \begin{cases} 
\frac{\hbar^2}{2m_L} k_x^2 + \frac{\hbar^2}{2m_L} k_y^2 - \Delta & \text{IF } d^\uparrow_{xy} \\
\frac{\hbar^2}{2m_L} k_x^2 + \frac{\hbar^2}{2m_L} k_y^2 & \text{bulk } d_{xy} \\
\frac{\hbar^2}{2m_L} k_x^2 + \frac{\hbar^2}{2m_H} k_y^2 & \text{bulk } d_{xz} \\
\frac{\hbar^2}{2m_H} k_x^2 + \frac{\hbar^2}{2m_L} k_y^2 & \text{bulk } d_{yz} 
\end{cases}
\]

Here, \(m_L\) stands for light mass while \(m_H\) stands for heavy mass. According to ARPES experiments\(^7\), \(m_L^* \approx 0.7m_e\), \(m_H^* \approx 10\sim20m_e\).

Without considering the spin degree of freedom, we can get the density of states (DOS) for the confined \(t_{2g}\) states:
\[
D = \begin{pmatrix}
\frac{m_L}{2\pi h^2} & d_{xy} \\
\sqrt{m_L m_H} & d_{xz} \\
\frac{2\pi h^2}{2\sqrt{m_L m_H}} & d_{yz}
\end{pmatrix}
\]

Taking the 3D conductivity tensor as a reference, we get the 2D conductivity tensor for the three \(t_{2g}\) states:

\[
\sigma^{xx} = \begin{pmatrix}
\frac{e^2\tau}{\pi h^2} \sigma_{IF}^{xy} & d_{xy} \\
\frac{e^2\tau}{\pi h^2} \frac{m_H}{\sqrt{m_L}} & d_{xz} \\
\frac{e^2\tau}{\pi h^2} \frac{m_L}{\sqrt{m_H}} & d_{yz}
\end{pmatrix}
\]

From this, we can see that the contribution of the \(d_{yz}\) state to \(\sigma^{xx}\) is very small compared that of the \(d_{xz}\) state.

With an external magnetic field \(H\), the majority band is shifted down and the minority band is shifted up. In our case, to first order, the change in the Fermi energy \(\delta \varepsilon_F\) could be obtained by number conservation:

\[
\delta \varepsilon_F \approx \frac{1}{2} g \mu_B H \frac{\sum t_{2g} D^\dagger(\varepsilon_F) - \sum t_{2g} D^\dagger(\varepsilon_F) - D^\dagger_{IF}(\varepsilon_F + \Delta)}{\sum t_{2g} D^\dagger(\varepsilon_F) + \sum t_{2g} D^\dagger(\varepsilon_F) + D^\dagger_{IF}(\varepsilon_F + \Delta)}
\]

where \(\sum t_{2g} D(\varepsilon_F)\) takes the summation of DOS over all three bulk \(t_{2g}\) bands at \(\varepsilon_F\) and \(D^\dagger_{IF}(\varepsilon_F + \Delta)\) refers to the DOS of the IF \(d^\dagger_{xy}\) band.

The conductivity without external magnetic field can be written as:

\[
\sigma^{xx}(H = 0) = \sigma_{IF,xy}^{xx} + \sigma_{bulk, d_{xy}(1)}^{xx} + \sigma_{bulk, d_{yz}(1)}^{xx} + \sigma_{bulk, d_{xz}(1)}^{xx} + \sigma_{bulk, d_{xz}(2)}^{xx} + \sigma_{bulk, d_{yz}(1)}^{xx} + \sigma_{bulk, d_{yz}(2)}^{xx} + \frac{e^2\tau_{IF}}{\pi h^2} \sigma_{IF}^{xy} \sigma_{IF}^{xy} + 2 \times \frac{e^2\tau_{bulk, d_{xy}}}{\pi h^2} \sigma_{IF}^{xy} \sigma_{IF}^{xy} + 2 \times \frac{e^2\tau_{bulk, d_{xz}}}{\pi h^2} \sigma_{IF}^{xy} \sigma_{IF}^{xy} + 2 \times \frac{e^2\tau_{bulk, d_{yz}}}{\pi h^2} \sigma_{IF}^{xy} \sigma_{IF}^{xy}
\]

\[
\frac{e^2\tau_{bulk, d_{yz}}}{\pi h^2} \sigma_{IF}^{xy} \sigma_{IF}^{xy} + 2 \times \frac{e^2\tau_{bulk, d_{xz}}}{\pi h^2} \sigma_{IF}^{xy} \sigma_{IF}^{xy} + 2 \times \frac{e^2\tau_{bulk, d_{yz}}}{\pi h^2} \sigma_{IF}^{xy} \sigma_{IF}^{xy}
\]
Under an external magnetic field $H$, the conductivity becomes:

$$\sigma_{xx}(H \neq 0) = \frac{e^2 \tau_{IF} \varepsilon_F + \Delta + \frac{1}{2} g \mu_B H}{2 \pi h^2} + \frac{e^2 \tau_{bulk \, d_{xy}} \varepsilon_F + \Delta \frac{1}{2} g \mu_B H}{\pi h^2} + \frac{e^2 \tau_{bulk \, d_{xz}} \varepsilon_F + \Delta \frac{1}{2} g \mu_B H}{\pi h^2} \sqrt{m_H \over m_L}$$

$$+ \frac{e^2 \tau_{bulk \, d_{yz}} \varepsilon_F + \Delta \frac{1}{2} g \mu_B H}{\pi h^2} \sqrt{m_L \over m_H}$$

By taking the difference and plugging in what we had before, we get:

$$\sigma_{xx}(H) - \sigma_{xx}(0) = \frac{e^2 \tau_{IF} \varepsilon_F + \Delta \frac{1}{2} g \mu_B H}{2 \pi h^2} \left( \frac{m_H}{2 \pi h^2} + 4 \frac{\sqrt{m_L m_H}}{2 \pi h^2} \right)$$

The change of magnetoresistance is:

$$\rho(H) - \rho(0) = \frac{1}{\sigma(H)} - \frac{1}{\sigma(0)} = \frac{1}{\sigma(0)} \left( \frac{1}{1 + \frac{\sigma(H) - \sigma(0)}{\sigma(0)}} - 1 \right) \approx - \frac{1}{\sigma(0)^2} (\sigma(H) - \sigma(0))$$

$$= - \frac{e^2 g \mu_B H}{2 \pi h^2} \left( \frac{m_L}{2 \pi h^2} + 4 \frac{\sqrt{m_L m_H}}{2 \pi h^2} \right)$$
To get a positive linear magnetoresistance (LMR), the sufficient conditions are:

\[
\tau_{IF} < \tau_{\text{bulk} \, d_{xy}}, \quad 2\tau_{IF} < \tau_{\text{bulk} \, d_{xz}} + \frac{m_L}{m_H} \tau_{\text{bulk} \, d_{yz}} \approx \tau_{\text{bulk} \, d_{xz}}
\]

Considering strong scattering at the interface, it is highly possible that the above conditions could be satisfied. Therefore, we argue that the Zeeman effect is the likely origin of positive LMR in our system.

**Supplementary Figures**

**Supplementary Figure S1**: XPS Eu (a) 4d and (b) 3d core level measurements for varied substrate temperature and oxygen partial pressure.
Supplementary Figure S2: (a) X-ray diffraction coupled scans with corresponding (b) in-plane reciprocal space map and positions in reciprocal space for (c) STO and (b) EuO with a 45° rotation of the surface unit cell.
**Supplementary Figure S3:** MLLS fitting of layer-resolved EELS spectra. Red is EuO and Green is Eu$_2$O$_3$. Only a small amount of Eu$^{3+}$ is present in the disordered region. (Reference spectra taken from Mairoser et al.\textsuperscript{9}).
Supplementary Figure S4: (a) Sheet resistance for a 7-nm EuO film as a function of temperature.
(b) Hall resistance $R_{XY}$ for a 7-nm EuO film at 120 K. Solid lines indicate linear fits.
Supplementary Figure S5: High-angle annular dark-field scanning transmission electron microscopy [110]-projection image of the EuO/STO interface.
Supplementary Figure S6: (a) To determine the valence band offset, the spectra for pure EuO (red shading) and STO (blue shading) were scaled, offset, and added together to create a simulated fit (purple line) to the measured valence band spectra for a heterostructure of 1 nm EuO on STO (black open circles). (b) Schematic of band alignment.
Supplementary Figure S7: The partial density of states projected onto Ti ions at each layer. Interface TiO$_2$ layer is at top and middle bulk TiO$_2$ layer is at bottom. $d_{xy}$ orbitals are marked as black line. $d_{xz}$ and $d_{yz}$ orbitals are marked as red line. $d_{3z^2-r^2}$ orbitals are marked as blue and $d_{x^2-y^2}$ orbitals are marked as magenta.
Supplementary Figure S8: Resonance photoemission spectra of valence band near Eu 3d threshold. We see a strong resonance of the Eu\(^{2+}\) states in the valence band at \(h\nu = 1128\) eV and of Eu\(^{3+}\) states at \(h\nu = 1130\) eV. No resonating states are seen at the \(E_F\).
Supplementary Figure S9

2DEG experimental band structure along the ΓX directions measured for s-polarization and two photon energies $h\nu = 460.3$ eV (upper row) and 466 eV probing $d_{xy}$- and $d_{yz}$-states (bottom row). Spectra were measured for different X-ray irradiation times: fresh spot; and ‘burnt” spot after 30 min irradiation of the sample surface. The SX-ARPES time evolution spectra show the gradual increase of the spectral intensity indicating the sample recovery.
Supplementary Figure S10

(a) Absorption spectra near Eu 4f edge. Spectra are measured at 2 spots of the sample surface: fresh spot and “burnt” spot. “Burnt” spot spectrum shows huge increase of the Eu$^{2+}$ ions in comparison to Eu$^{3+}$. (b) Valence band spectrum measured at $h\nu = 457$ eV at “burnt” spot of the sample surface (blue line) shows the prevalence of Eu$^{2+}$ in comparison to the spectrum measured at fresh spot (green line).

Supplementary References

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